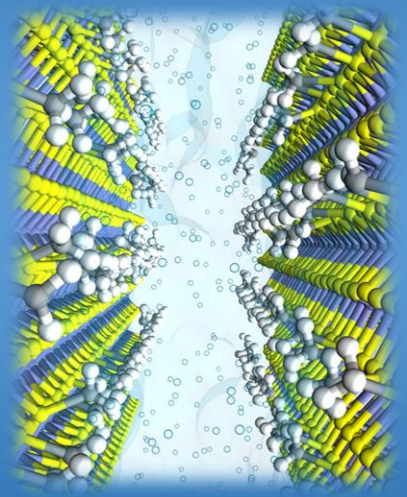
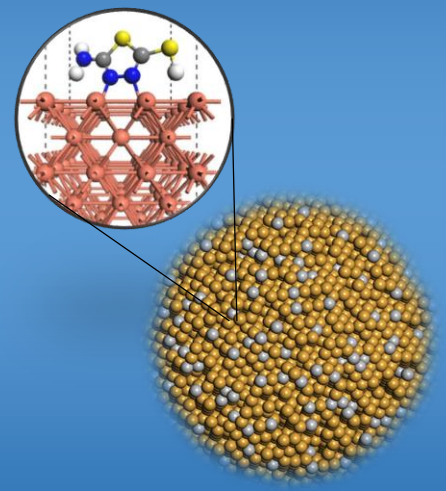
Nanochemistry on low-dimensional materials for electrocatalytic CO_2 conversion and water depollution



Damien VOIRY

Institut Européen des Membranes, UMR 5635 CNRS/ENSCM/UM

damien.voiry@umontpellier.fr
Website: lowdimensional-materials.net/
Twitter: @VoiryGroup



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  - Rafael Silva (Maringa State Univ., Brazil)
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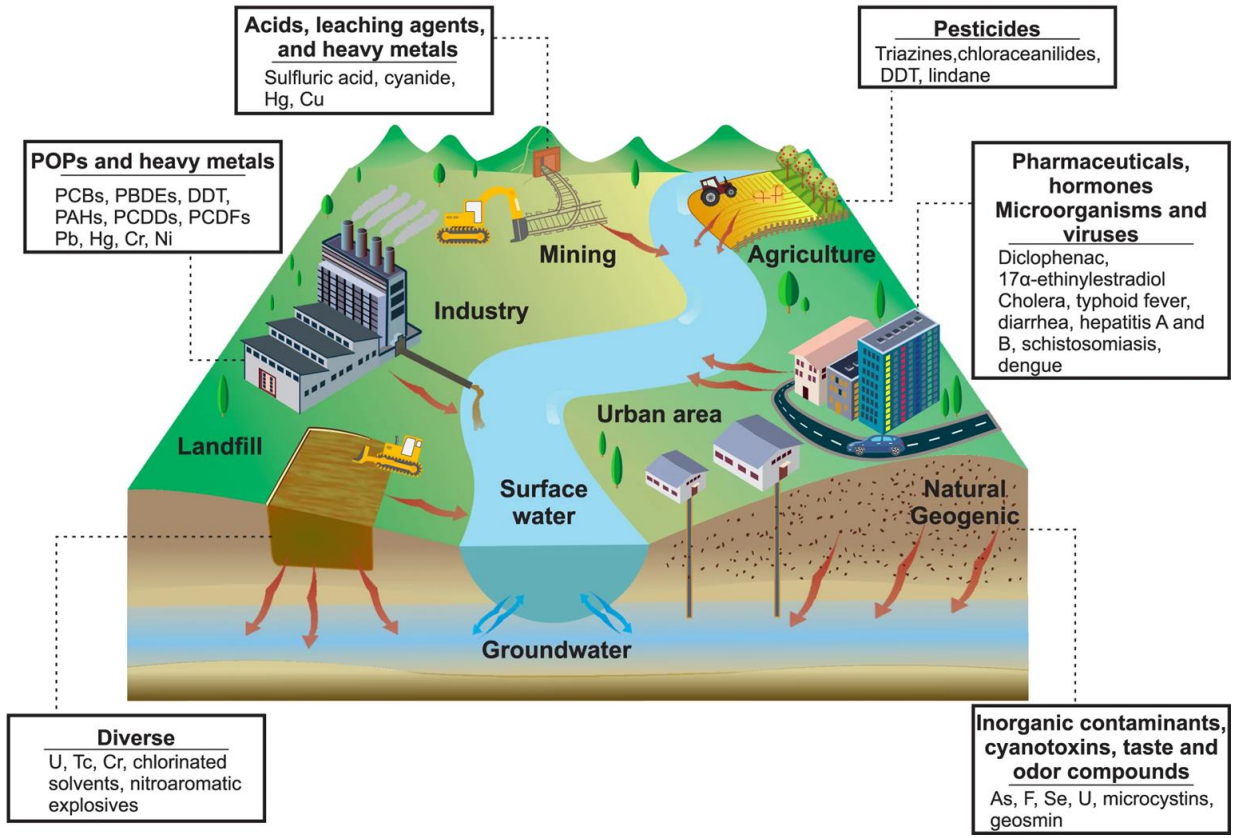
- Fundings

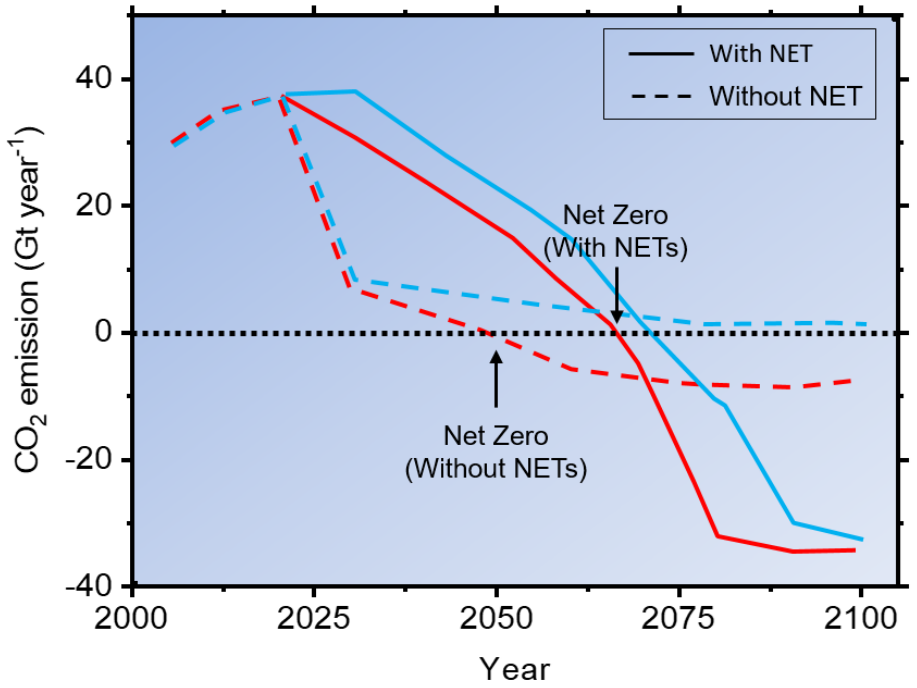
World challenges

RECLAIMING WATER



Peydayesh & Mezzenga, ***Nature Communnications****, 2021*

CO_2 RECYCLING



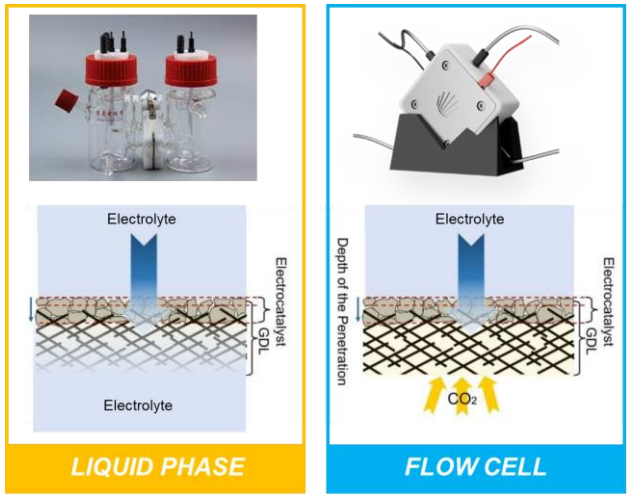
Adated from Realmonte, G.. et al.
Nature Communications*, 2019*

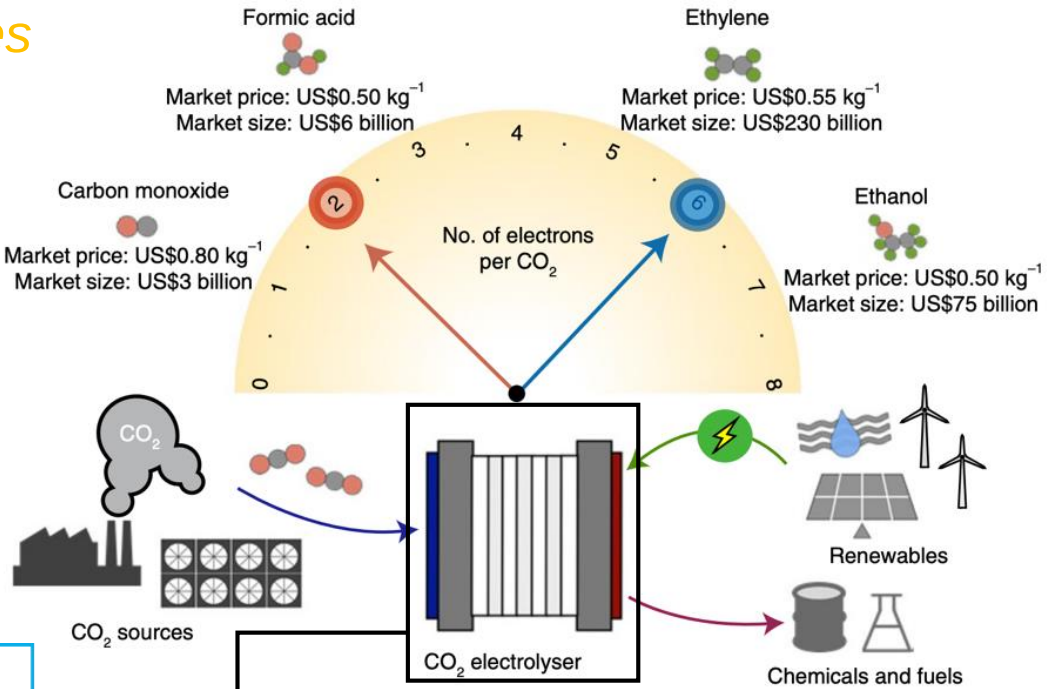
GLOBAL PROBLEMS → FUNDAMENTAL APPROACHES

CO_2 electrochemical reduction as NET?

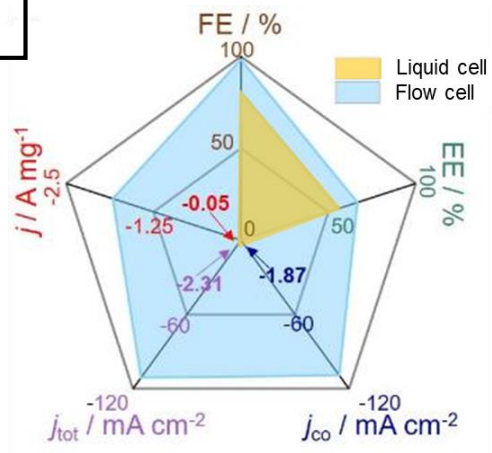
- *Techno-economic analyses*

- ***40 GT** of CO_2 per year*
- C_2H_4: **0.6 %** of the CO_2 emission
- Large market size: 230 B$



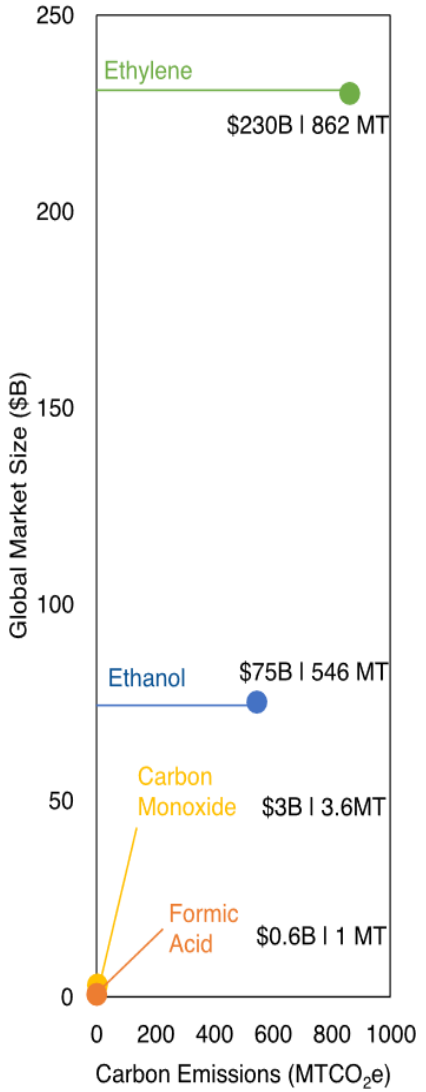


Shin et al. ***Nature Sust.*** 2021

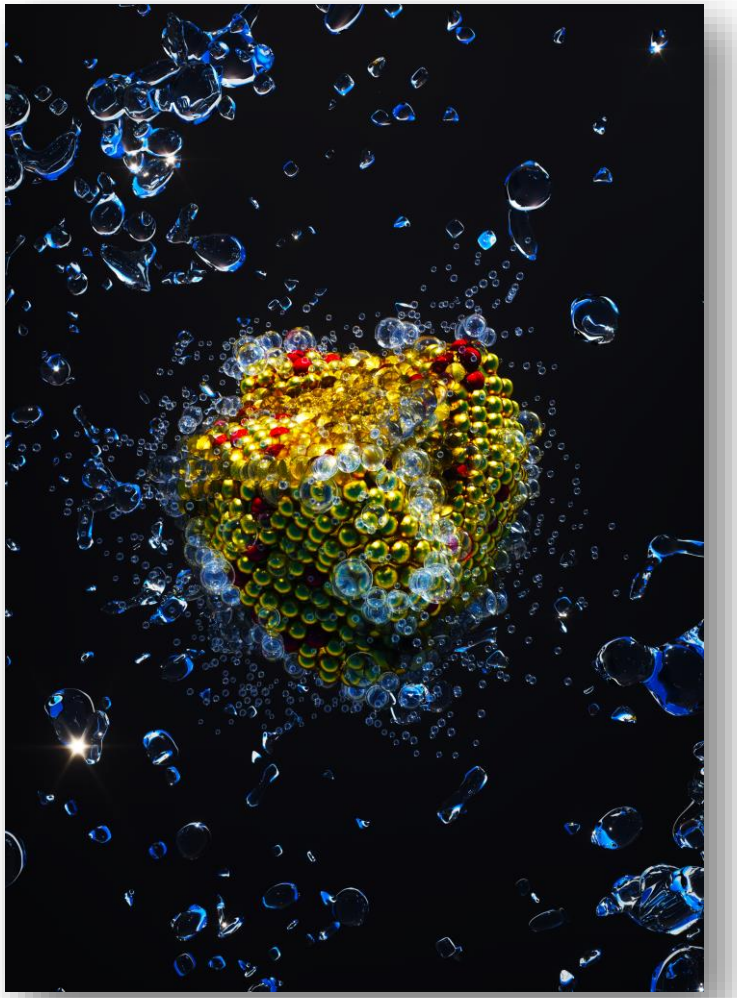


Zhang et al. ***ACS Catalysis.*** 2021

De Luna et al. ***Science.*** 2019



Towards more than 2 carbon products?

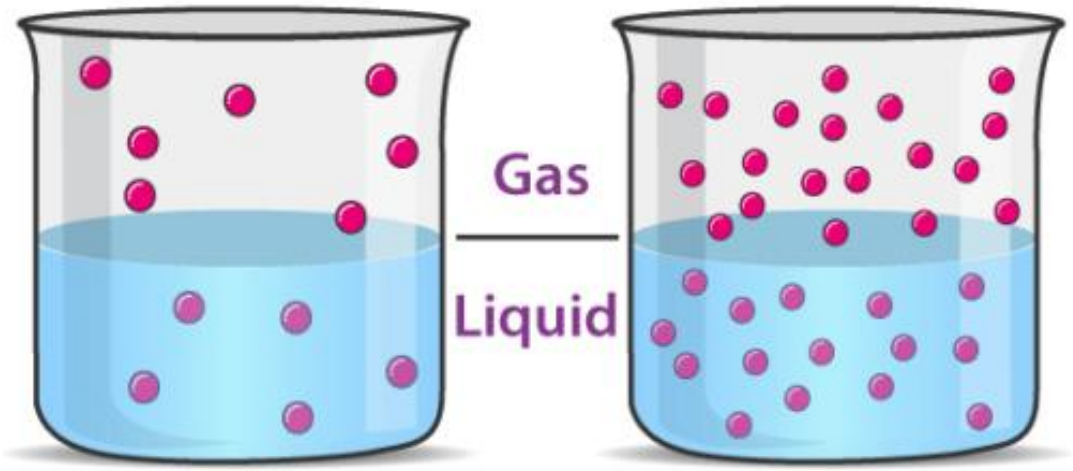


Design of new Cu-based alloy


Kun Qi


Nicolas Onofrio

Henry law: Under constant temperature the amount of a given gas that dissolves in a given volume of liquid is directly proportional to the partial pressure of gas in equilibrium with the liquid



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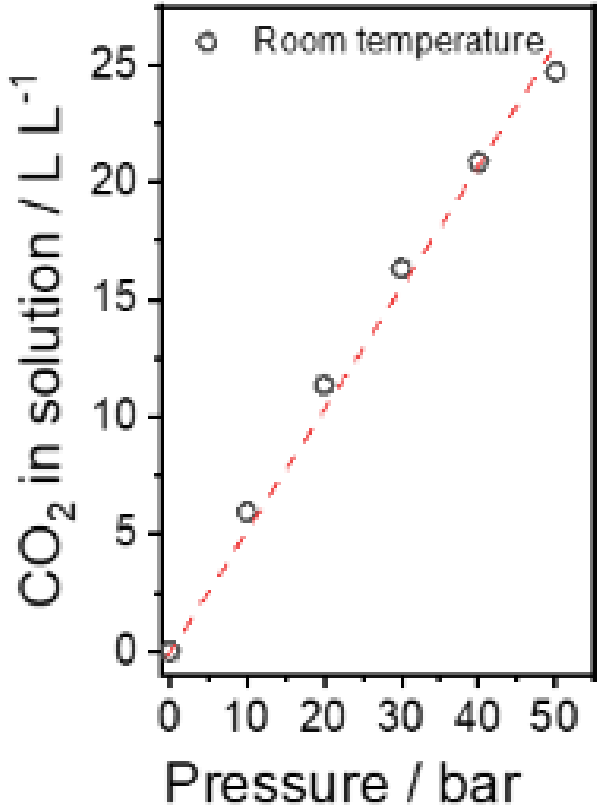
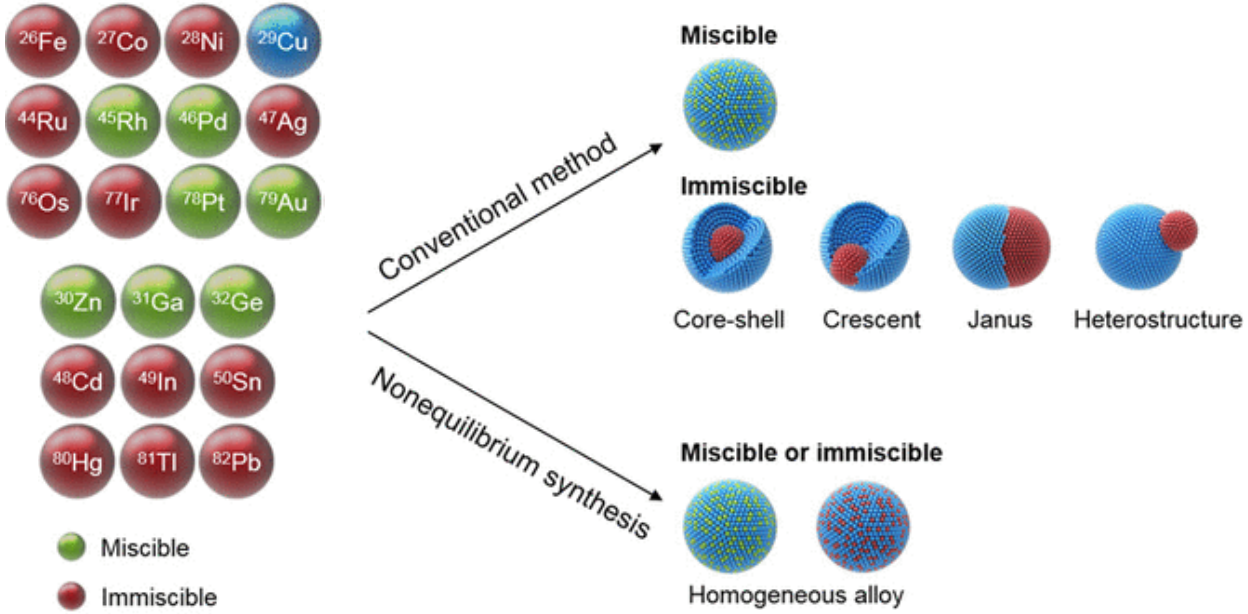
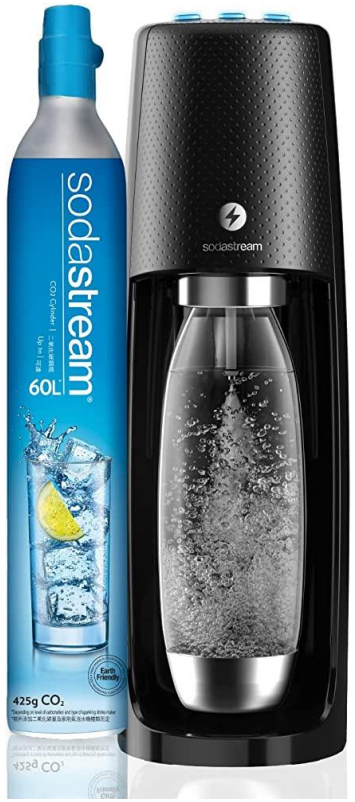
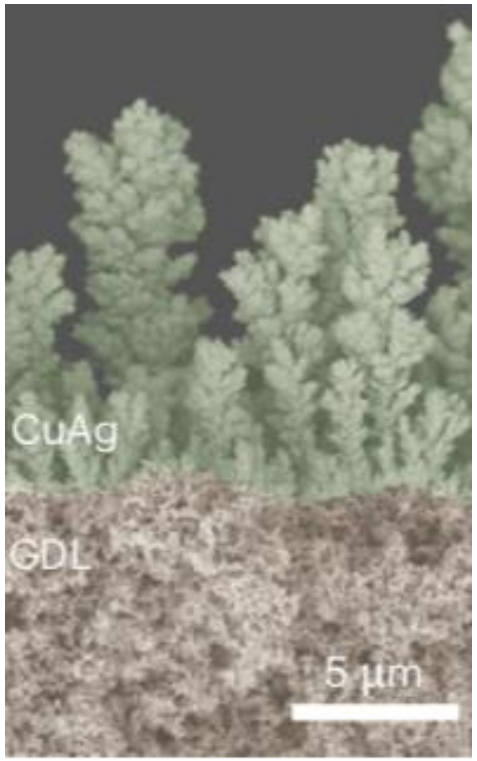


Design of new Cu-based alloy

***Electrosynthesis under supersaturated conditions*:**



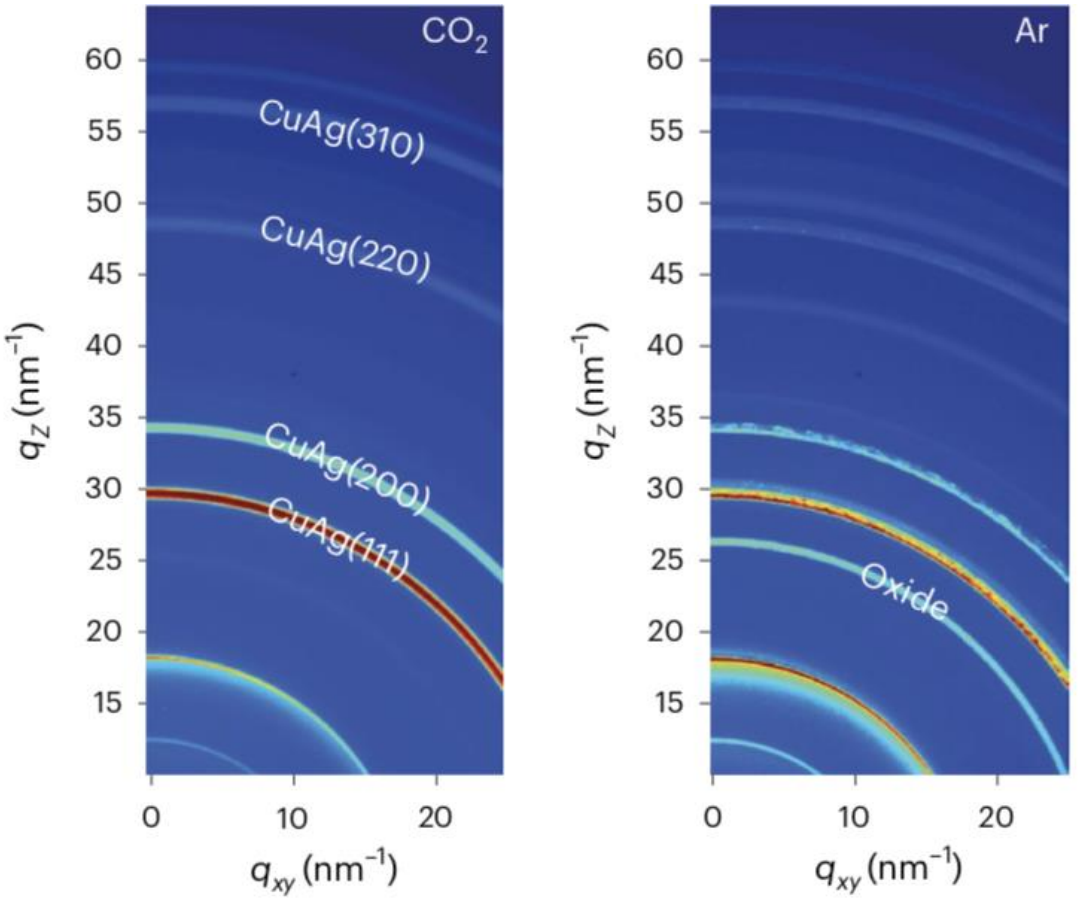


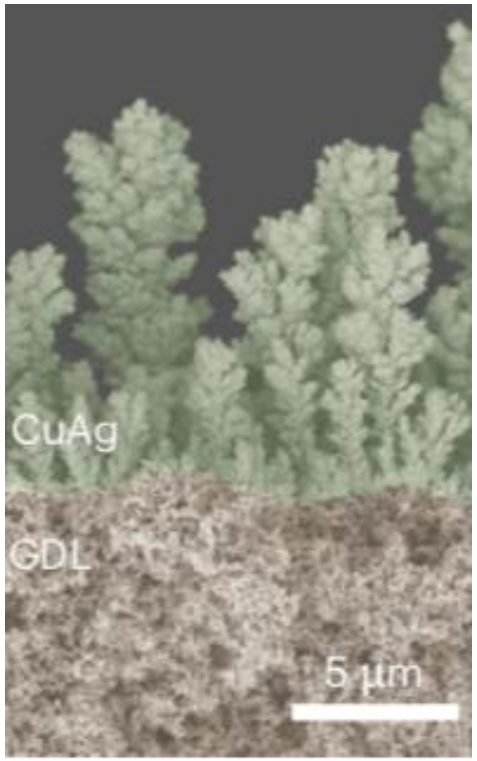


Qi, K. *et al.* **Nature Catalysis,** 2023

Design of new Cu-based alloy

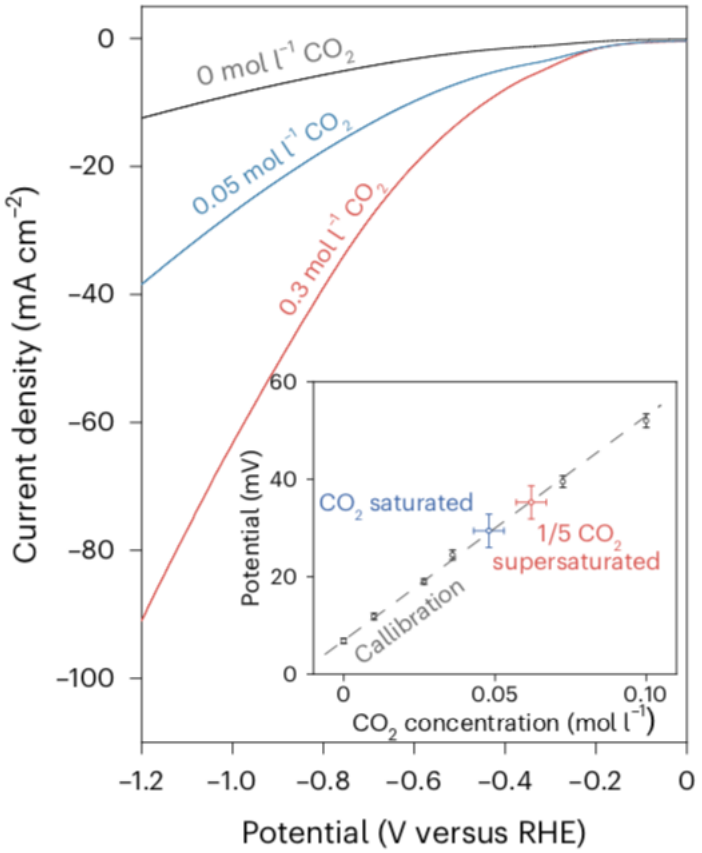
***Electrosynthesis under supersaturated conditions*:**

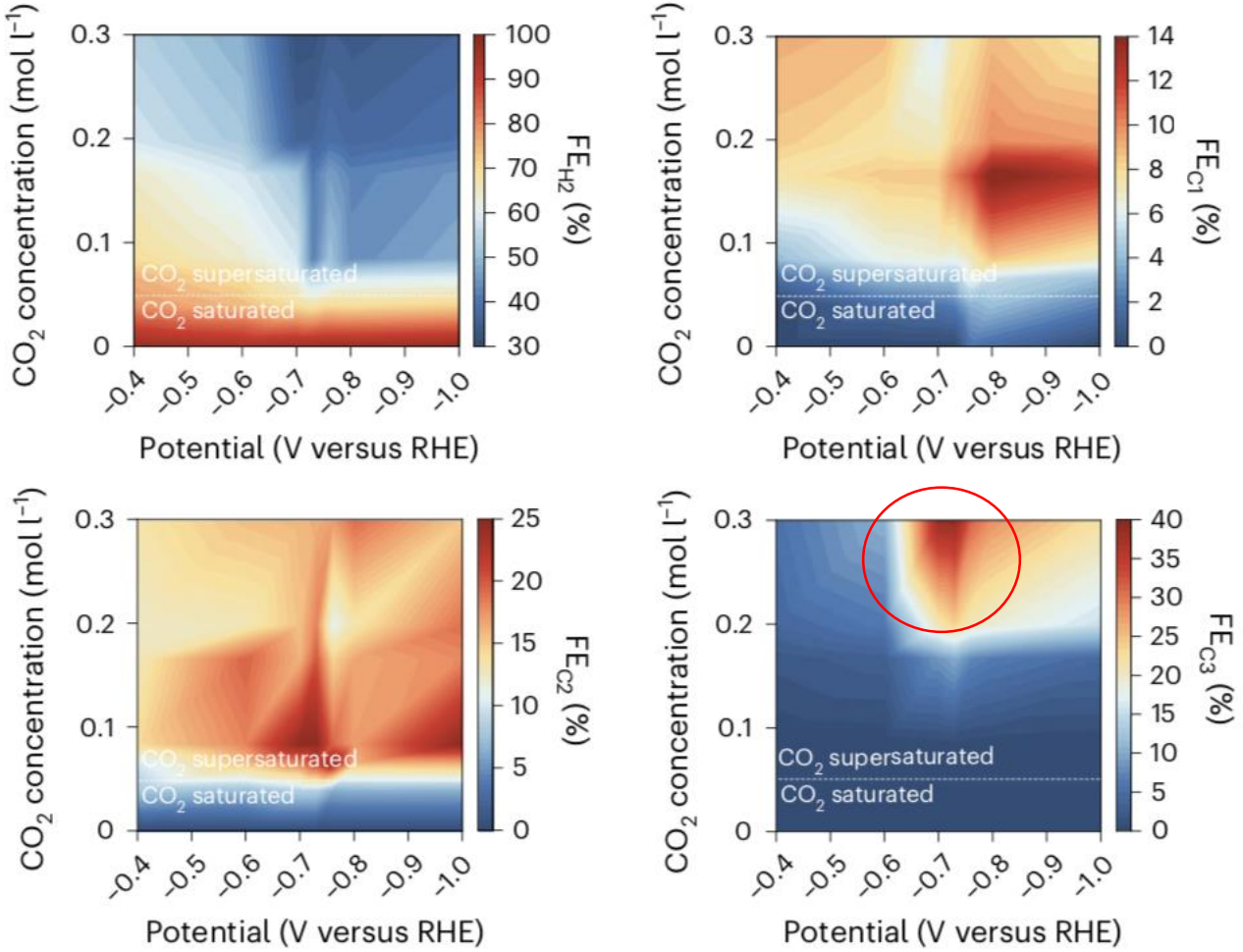




Qi, K. *et al.* **Nature Catalysis,** 2023

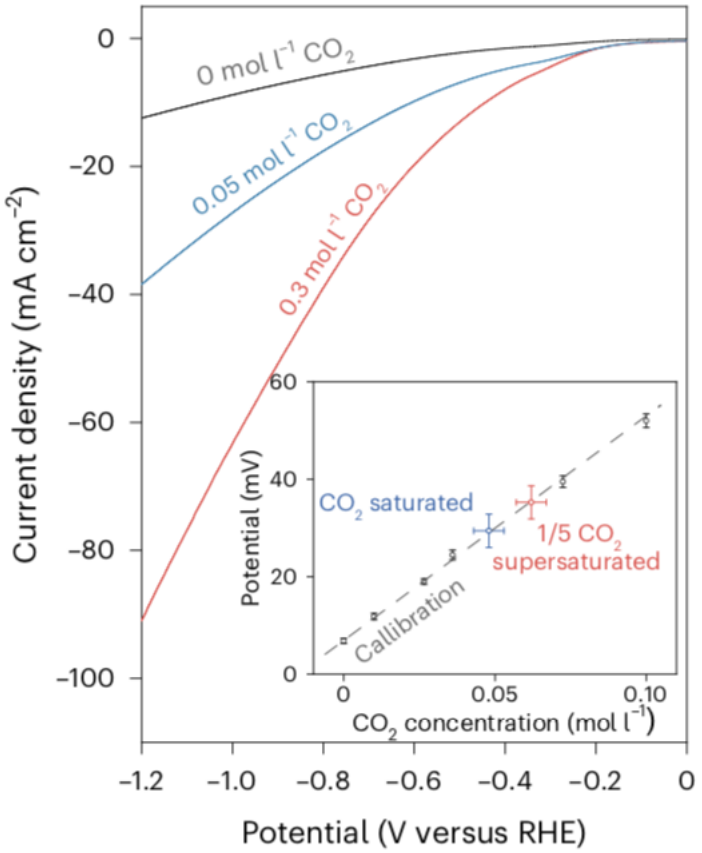
Electro-conversion of CO_2 to C_3

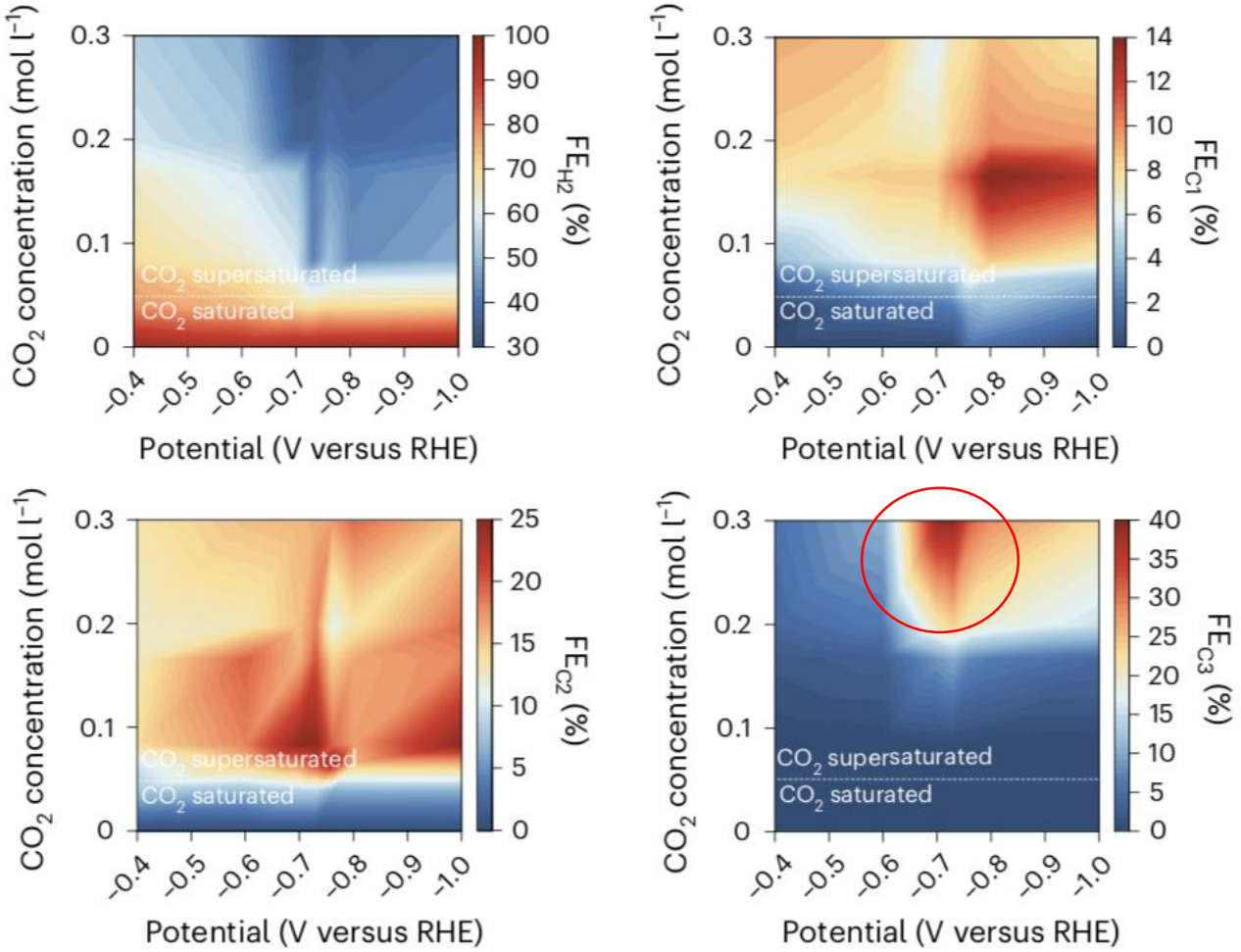




Qi, K. *et al.* **Nature Catalysis,** 2023

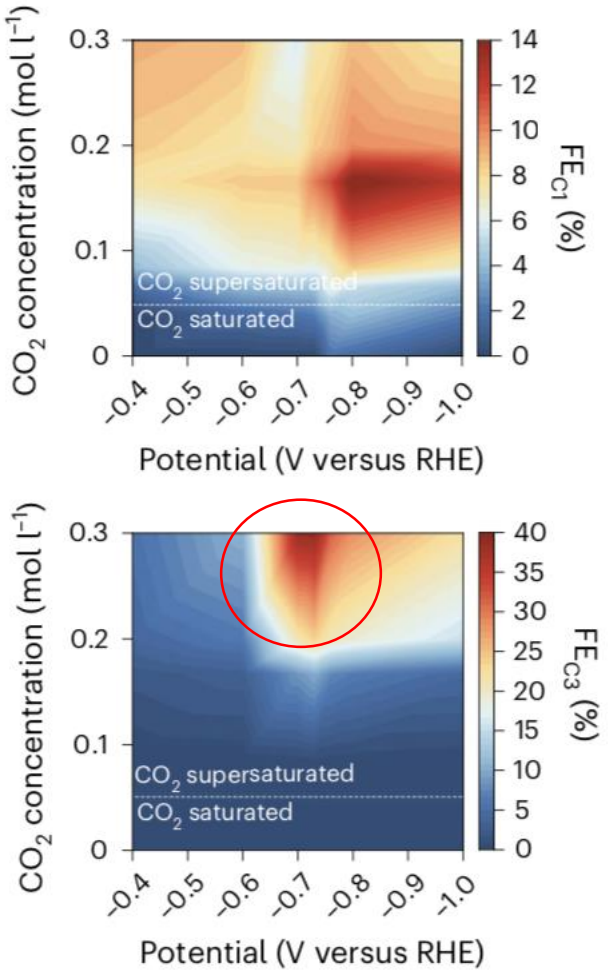
Electro-conversion of CO_2 to C_3





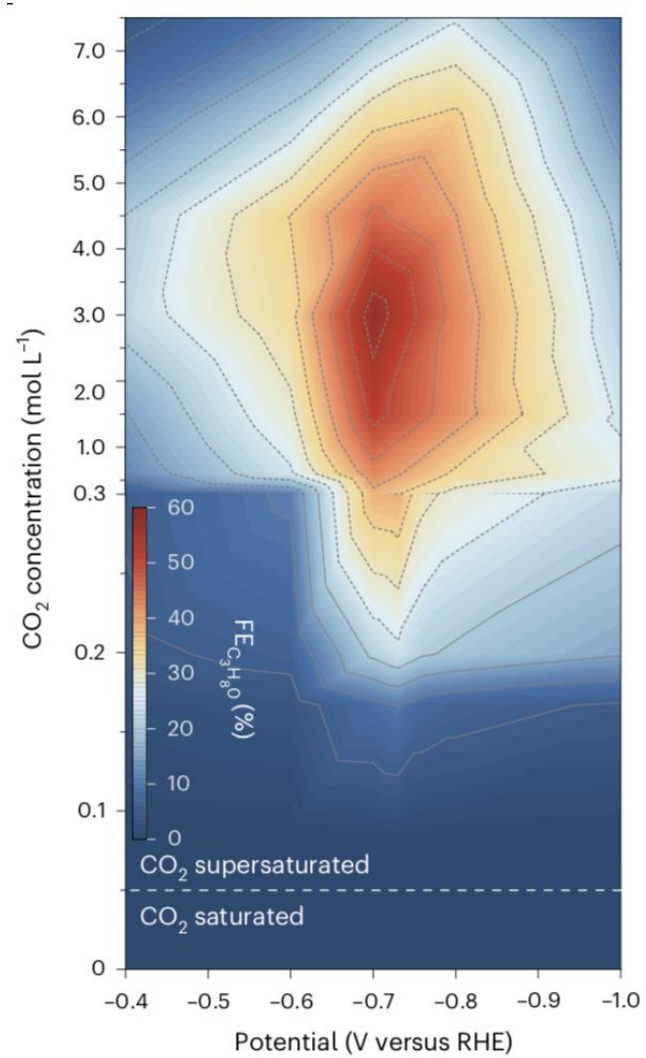
Qi, K. *et al.* **Nature Catalysis,** 2023

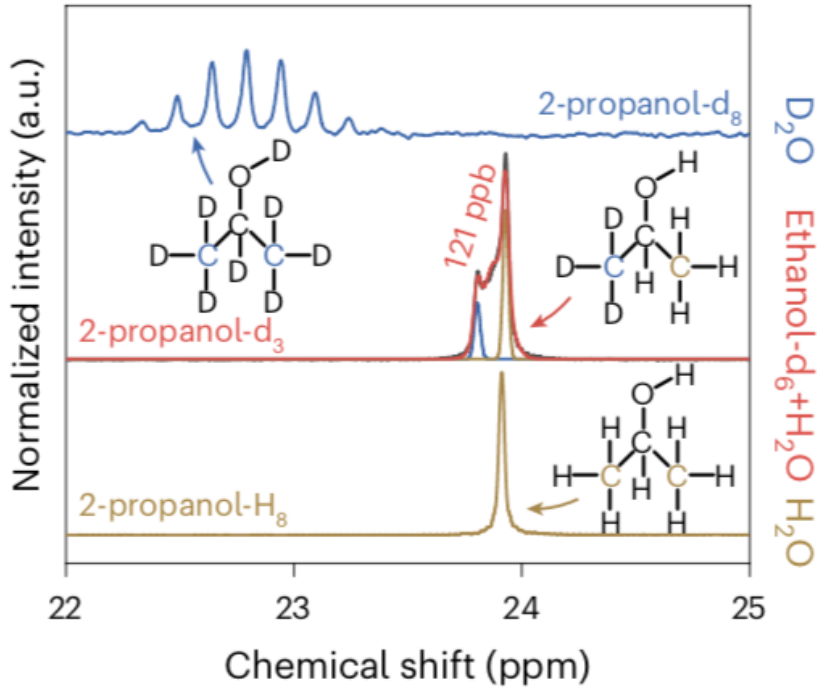
Electro-conversion of CO_2 to C_3



Qi, K. *et al.* **Nature Catalysis,** 2023

Electro-conversion of CO_2 to C_3



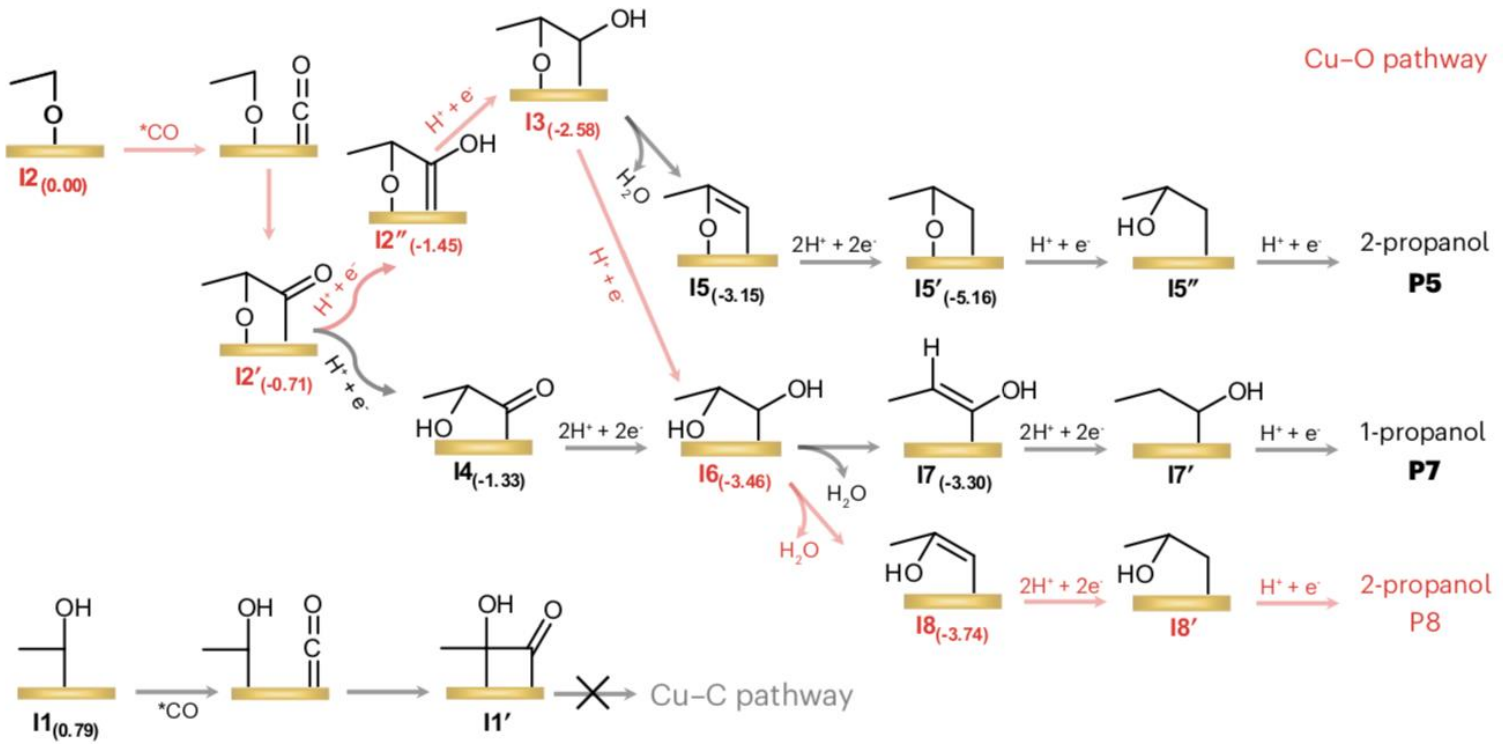


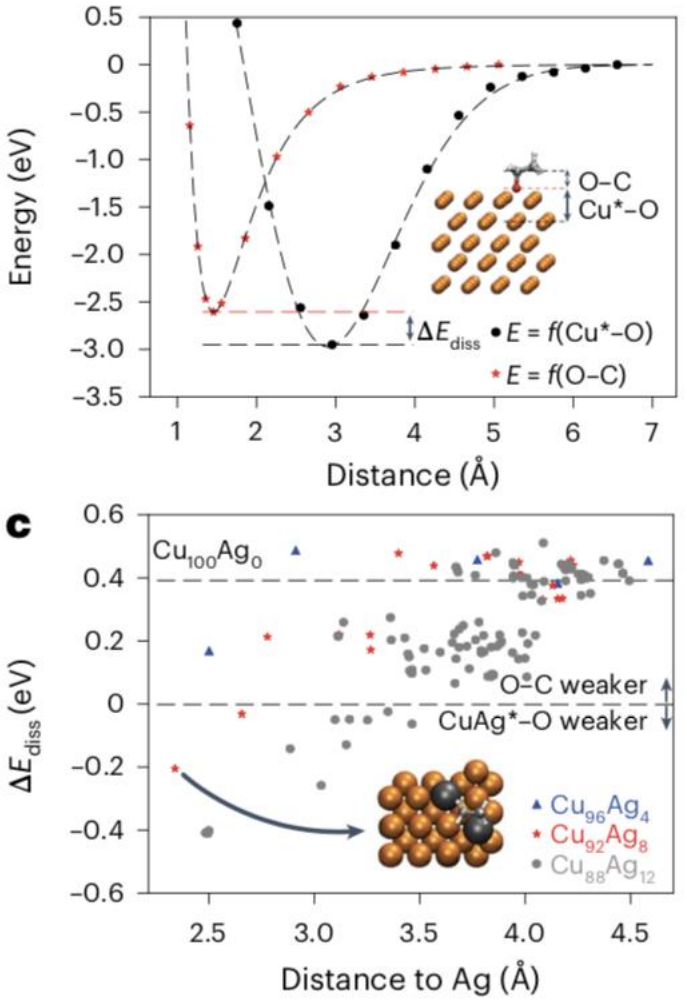
- Production of 2-propanol with FE ~ 57%
- Confirmed by isotopic labeling experiements

Qi, K. *et al.* **Nature Catalysis,** 2023

Electro-conversion of CO_2 to 2-propanol

Mechanistic investigations

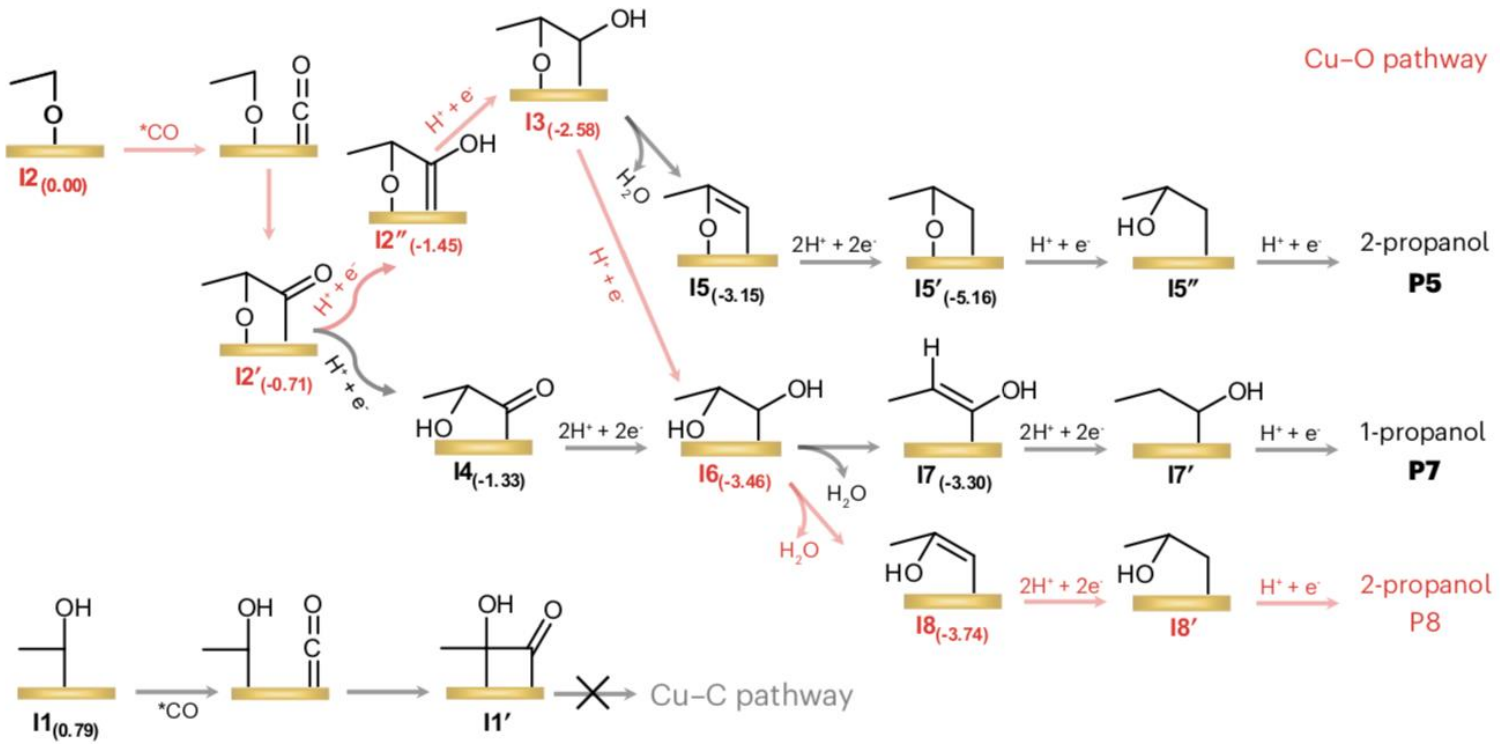


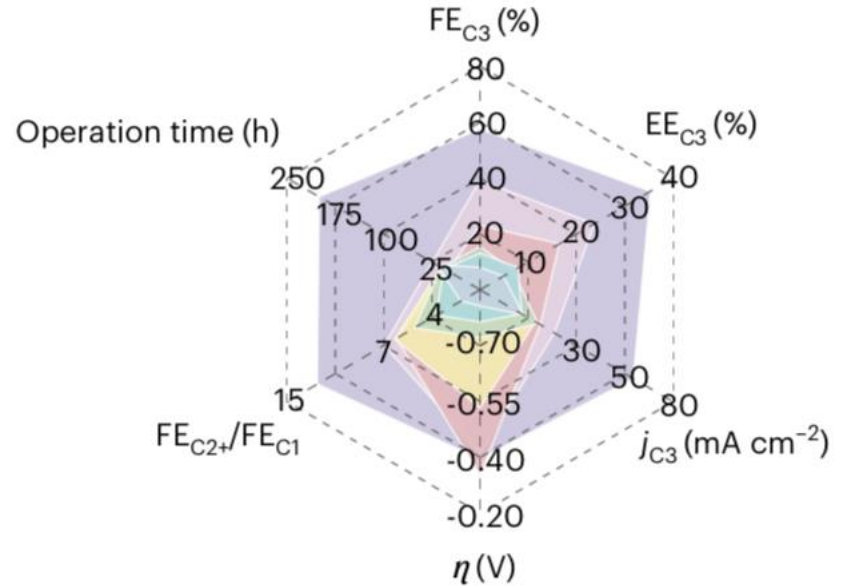


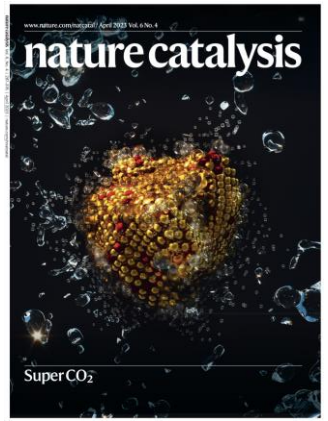
Qi, K. *et al.* **Nature Catalysis,** 2023

Electro-conversion of CO_2 to 2-propanol

Mechanistic investigations



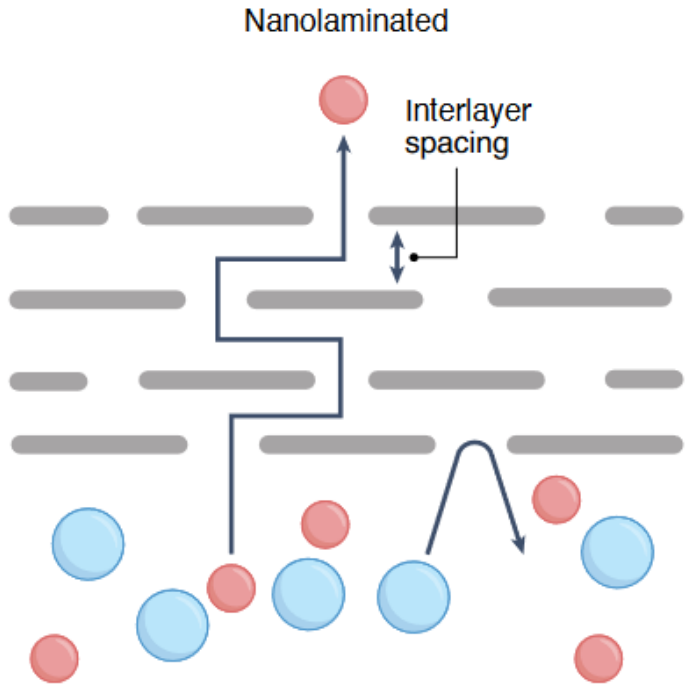




- Ag destabilizes Cu-O bonds
- High selectivity for 2-propanol

Qi, K. *et al.* **Nature Catalysis,** 2023

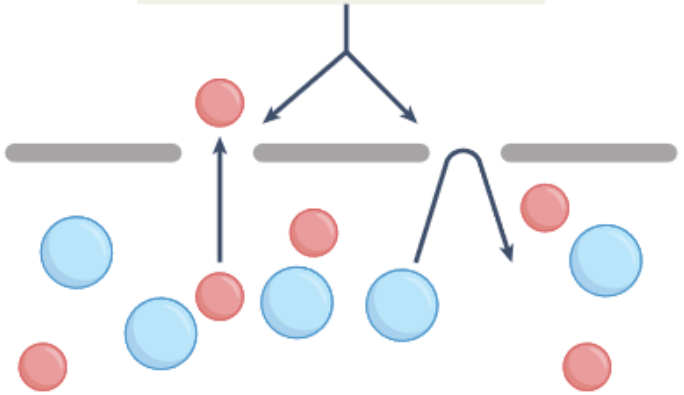
Molecular sieving from 2D materials

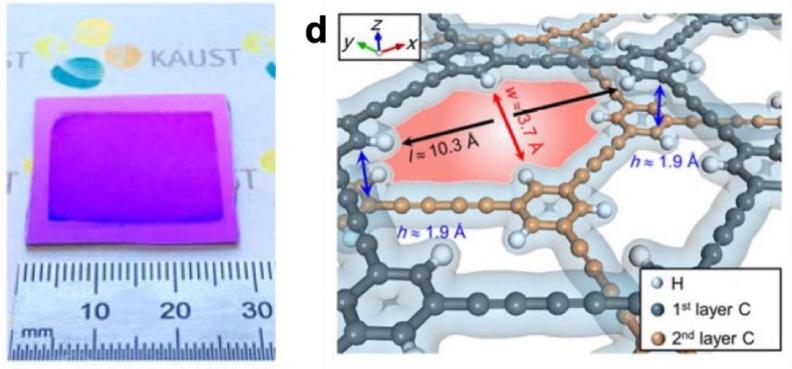


Zoology of 2D-based membranes

Single layer

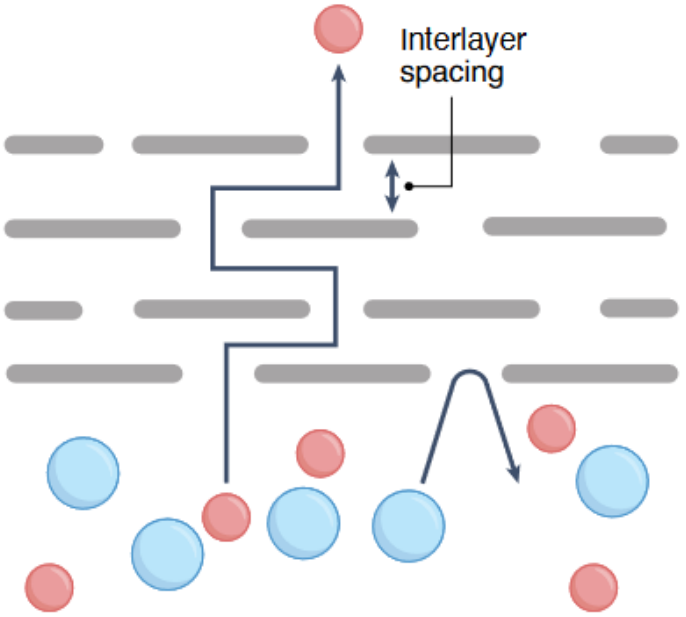
Nanopores:
- Extrinsic: defects
- Intrinsic: porous framework

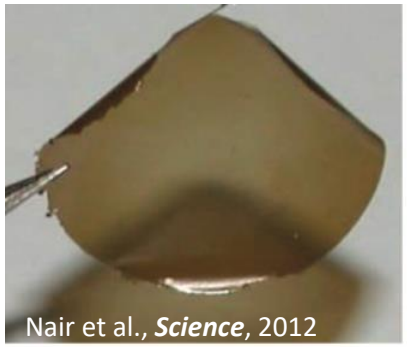




Shen et al., ***Nature Materials***, 2022

Nanolaminated



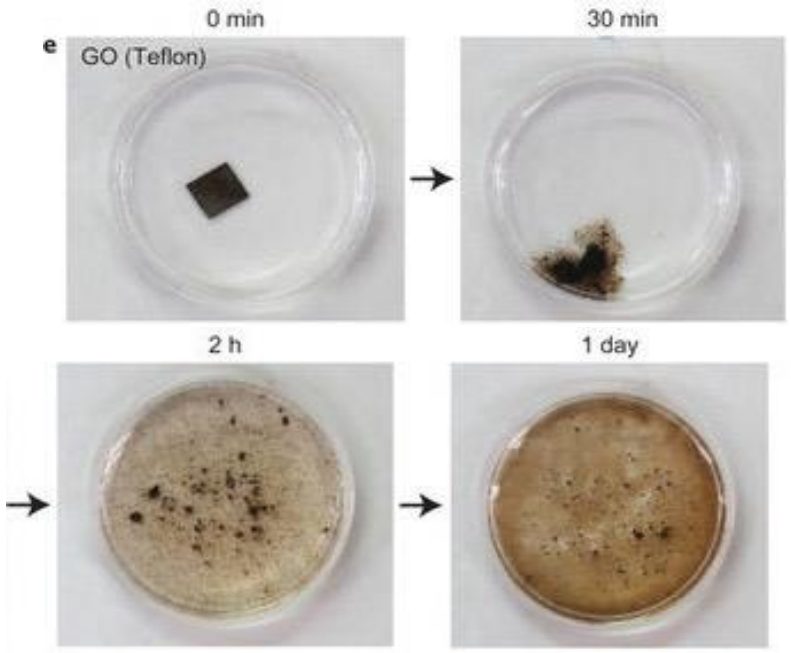


Nair et al., ***Science***, 2012

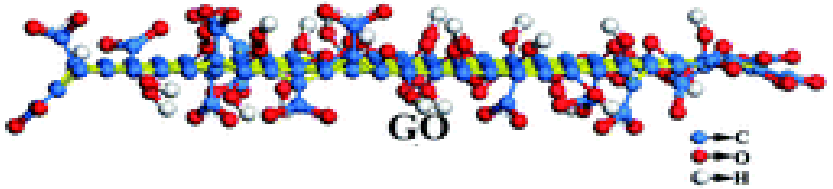
Salameh et al., ***Nature Materials***, 2022

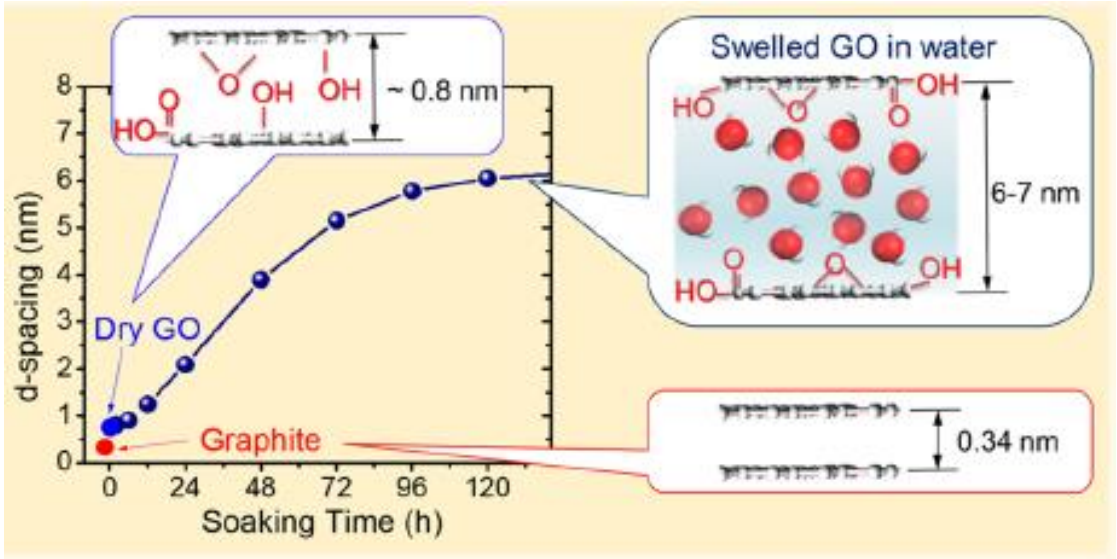
2D nanolaminate membranes

- *Graphene oxide membranes*



Yeh, Nat. Chem., 2015



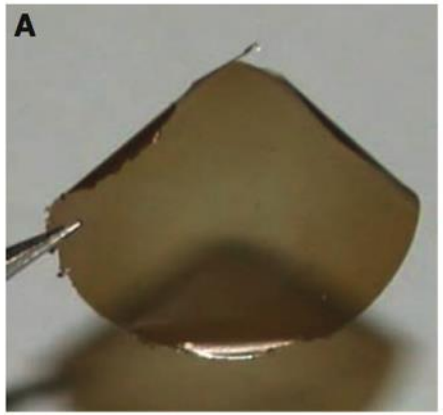


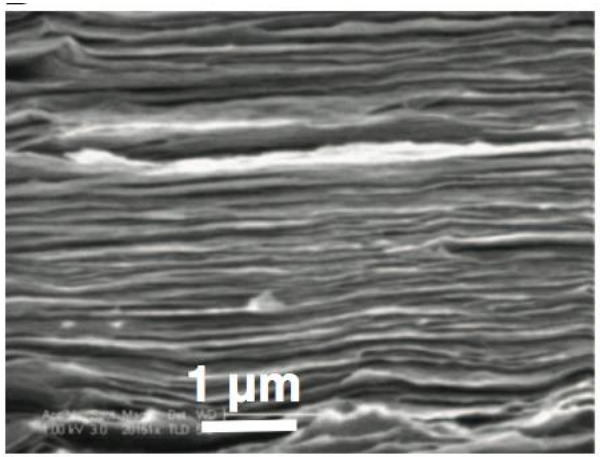
Zheng et al., ACS Nano, 2017

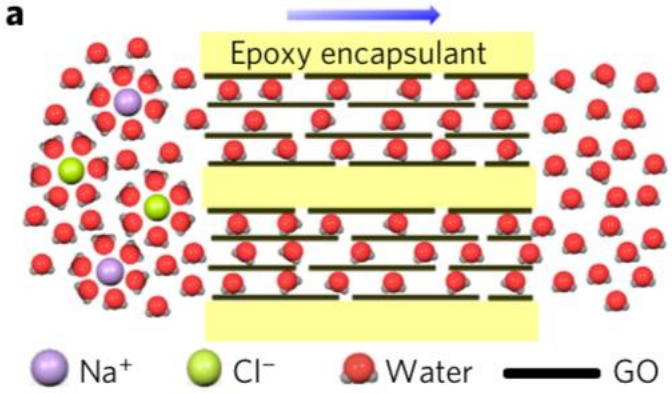
- Uncontrollable sweeling
  - Increasing of the membrane prosity
  - Loss of the physical integrity

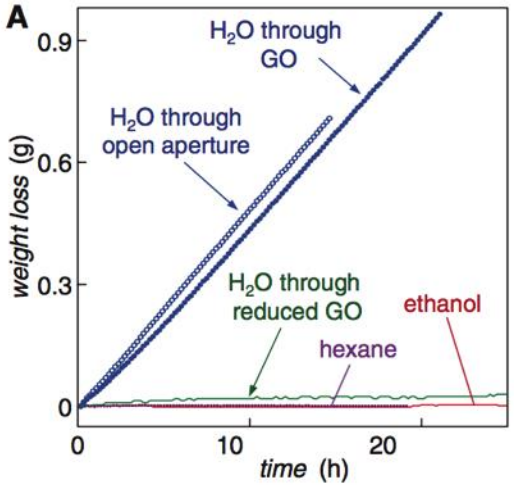
2D nanolaminate membranes

- *Graphene oxide membranes*

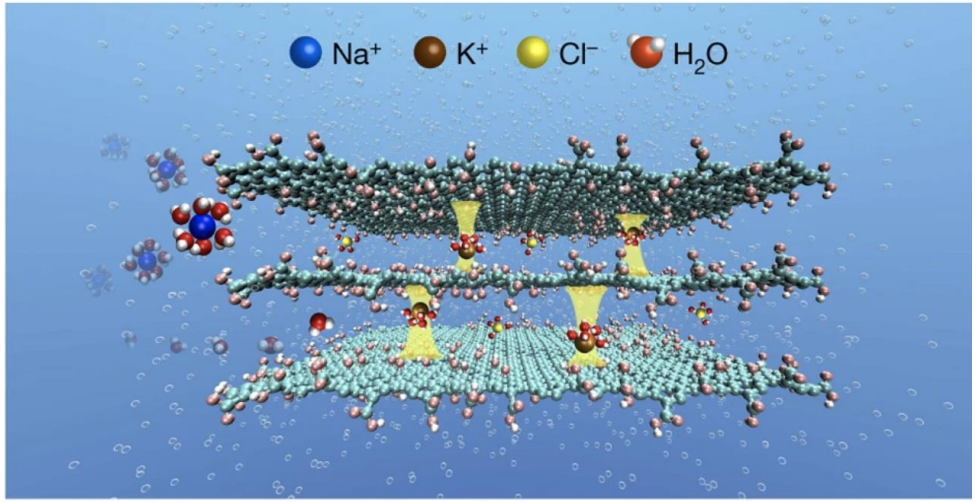




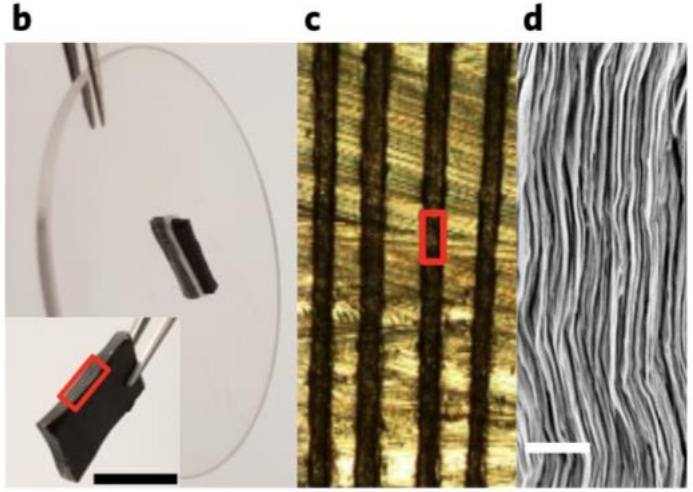




Nair et al., **Science**, 2012



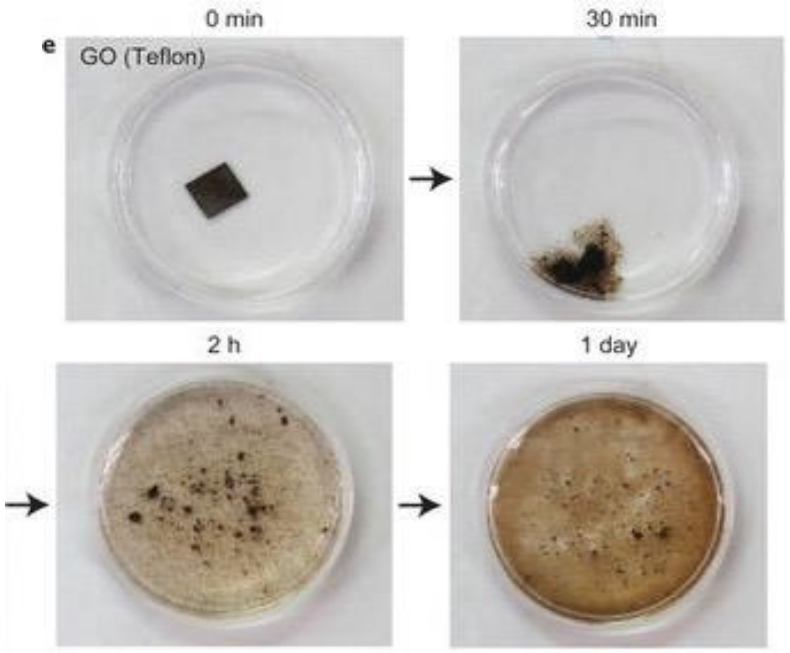
Chen et al., **Nature**, 2017



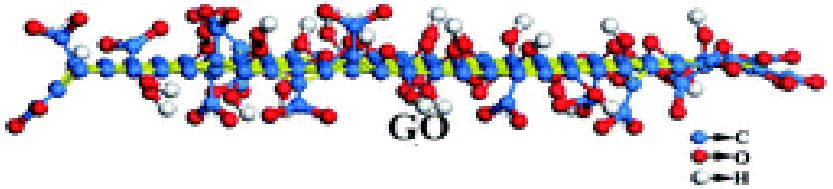
Abraham et al., ***Nature Materials***, 2017

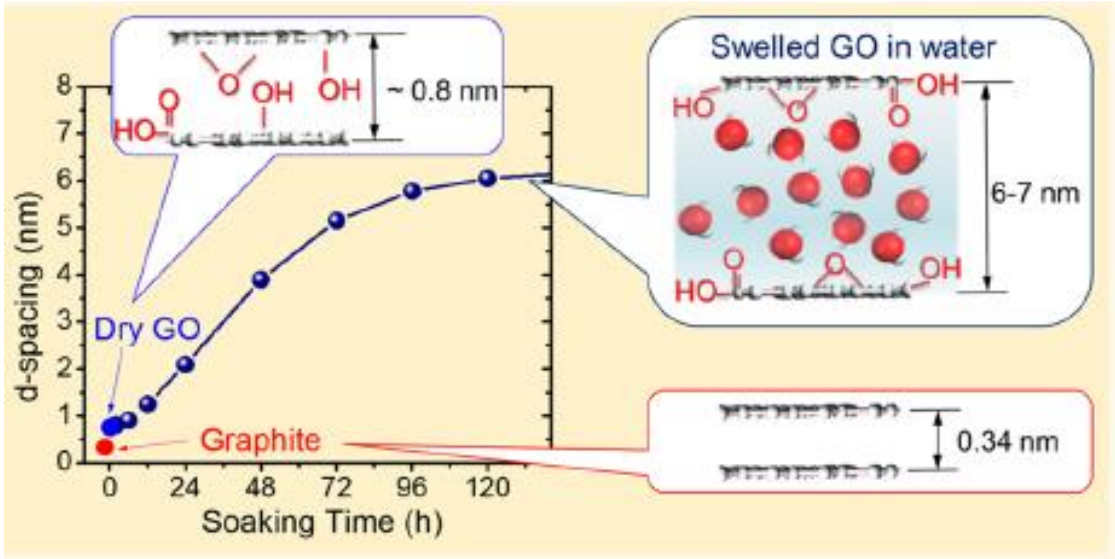
2D nanolaminate membranes

- *Graphene oxide membranes*



Yeh, Nat. Chem., 2015

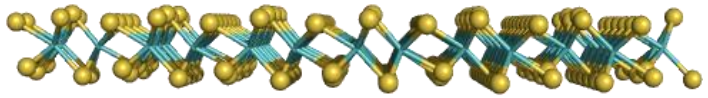




Zheng et al., **ACS Nano**, 2017

☹ Uncontrollable sweeling

- Molybdenum disulfide (MoS_2)



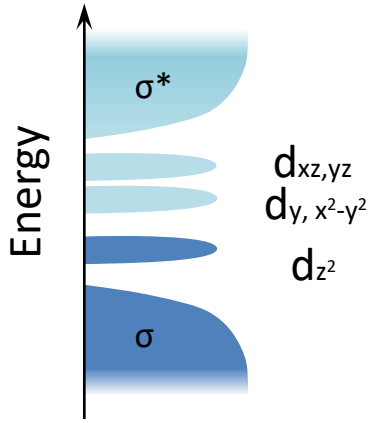
☹ Water impermeable when dried

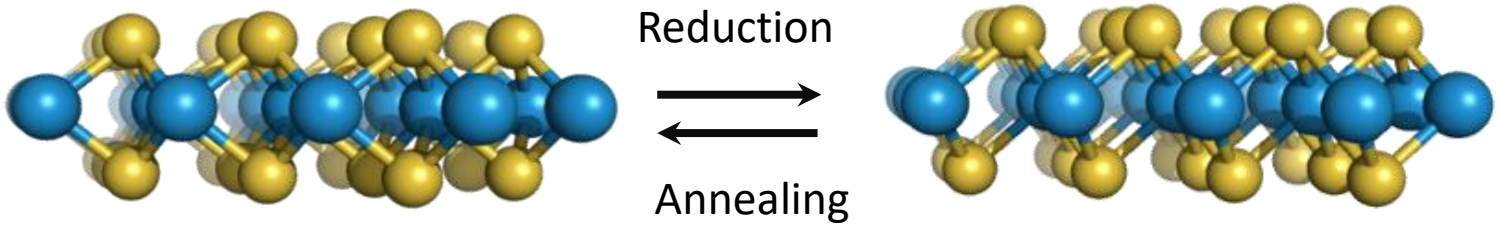
Wang et al., **Nano Lett**, 2017

Chemical exfoliation of group-6 TMDs (MX_2)

- *Intercalation-assisted exfoliation*

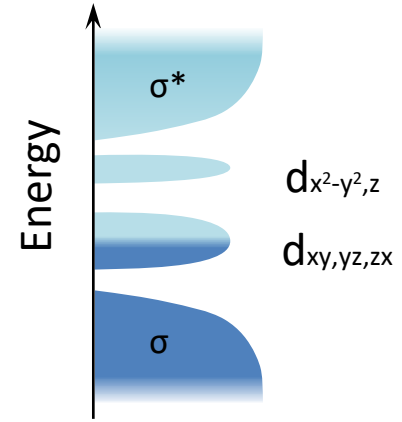
Joensen et al. ***R. Mater. Res. Bull.***, 1986

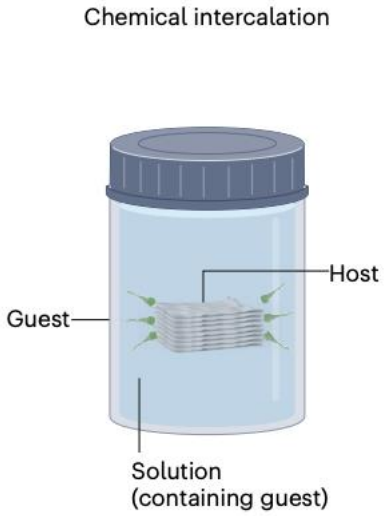


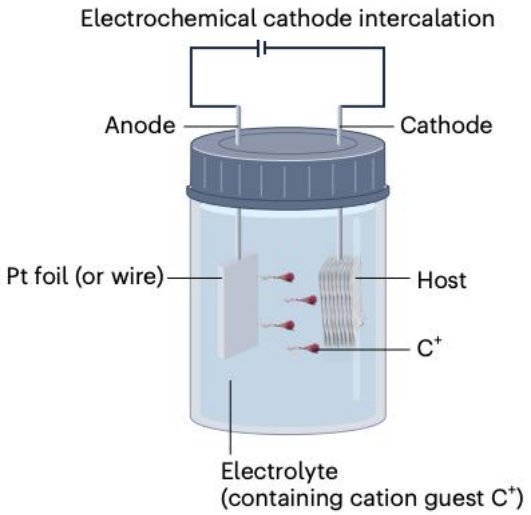


2H-MX_2: semi-conductor

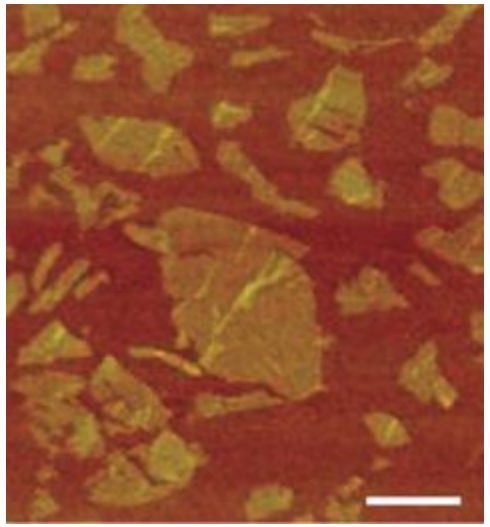
1T-MX_2: metallic

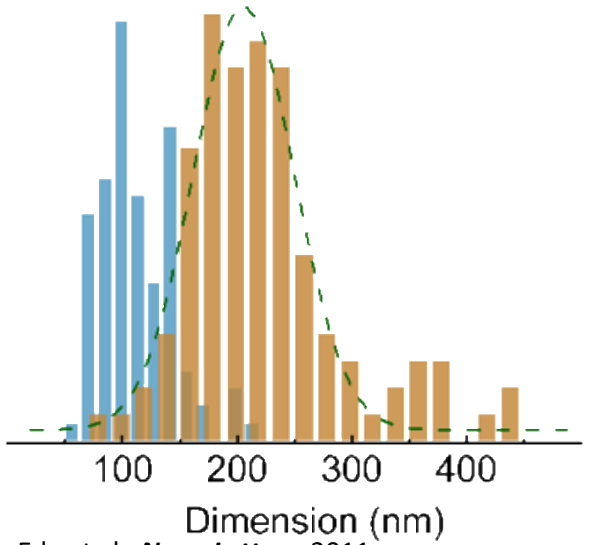






Yang et al., ***Nature Synthesis***, 2023
Yang et al., ***Nature Protocols***, 2022





Eda et al., ***Nano Letters***, 2011

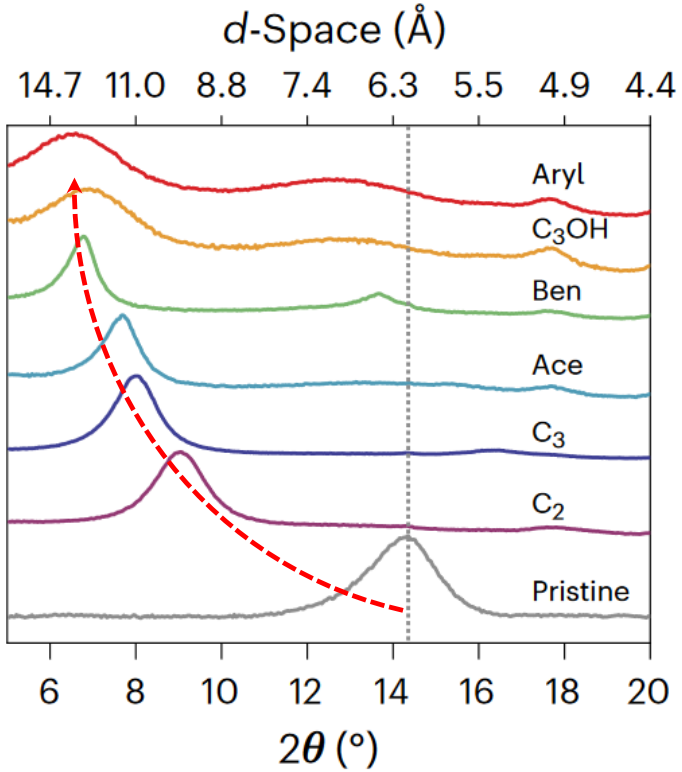
Role of stacking disorders

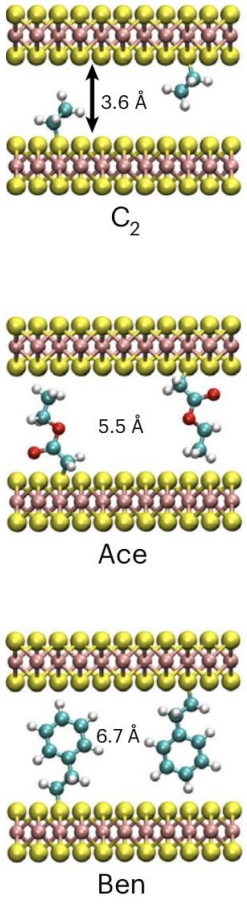
Wensen Wang

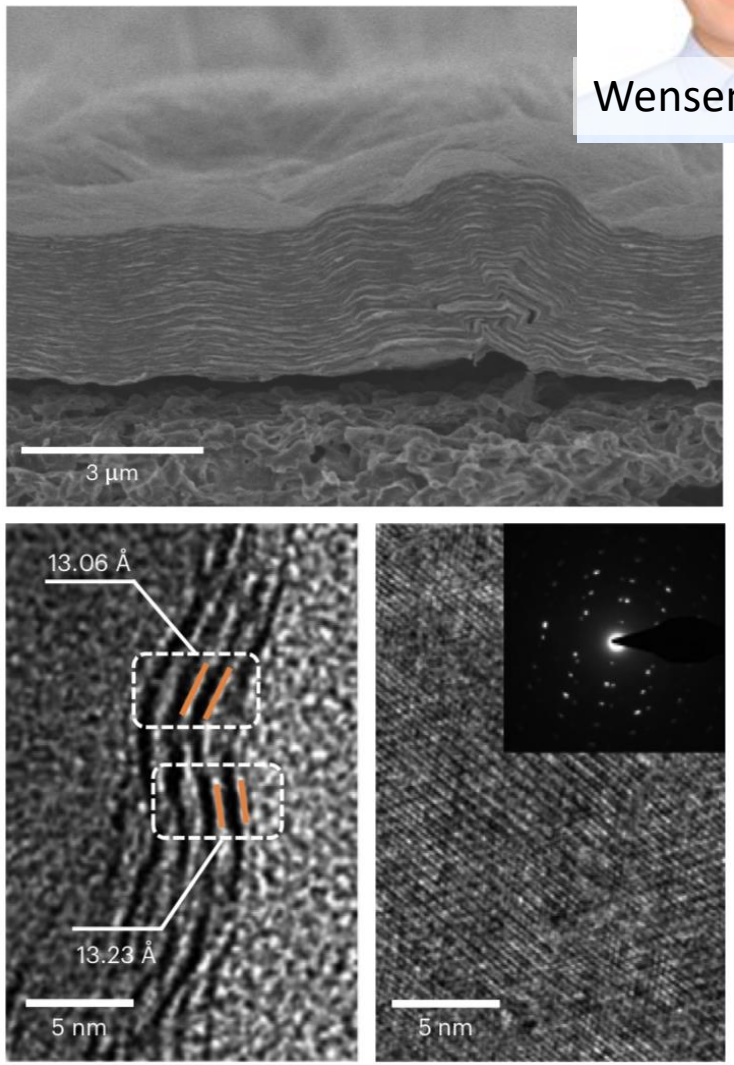
- *Development of novel functionalization chemistry*

  - Wider control of the interlayer space via covalent functionalization

  - Tunable stacking disorder of the nanosheets



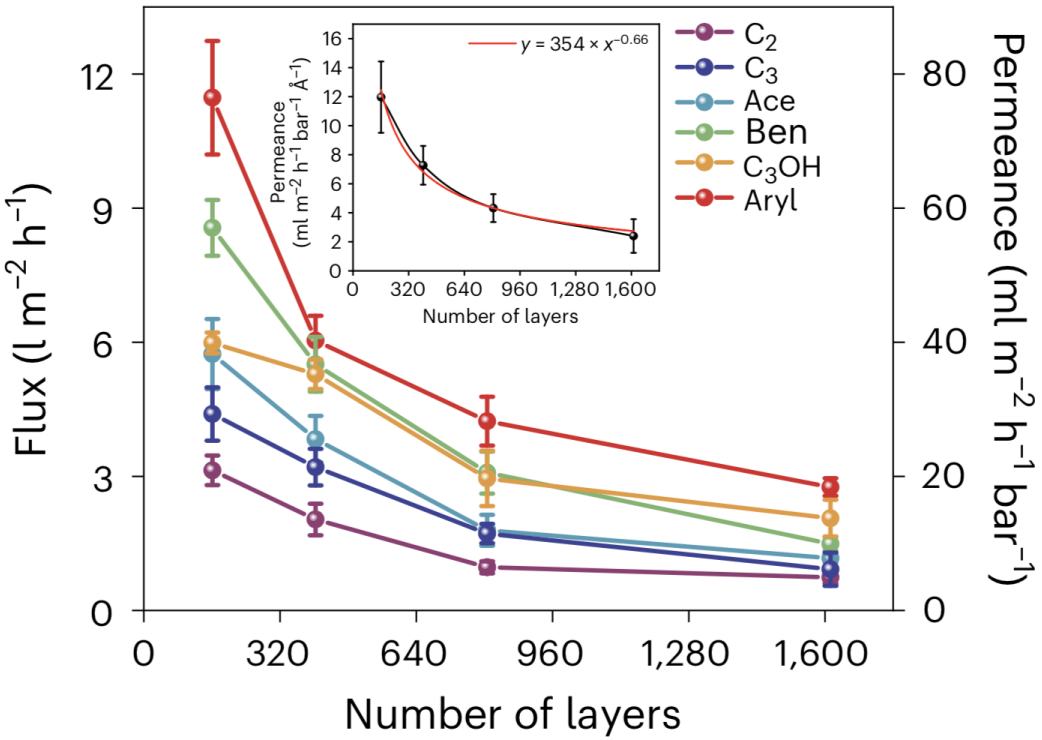


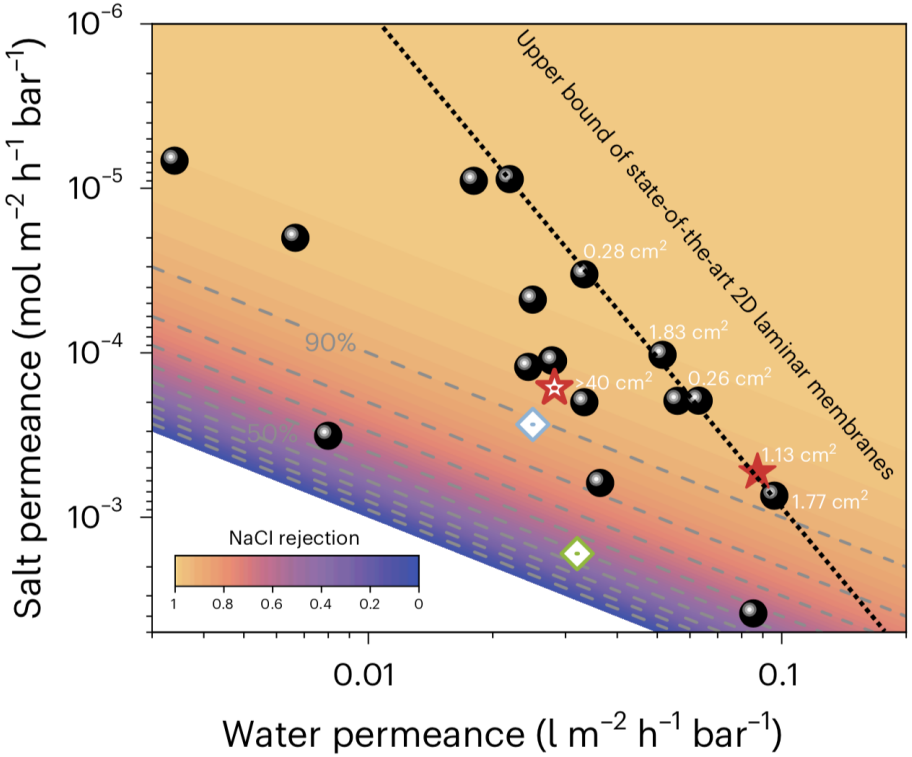


Wang et al., ***Nature Water****, 2023*
See also: Ries et al. ***Nature Materials****, 2019*

Role of stacking disorders

- Nanolaminate membranes for forward osmosis



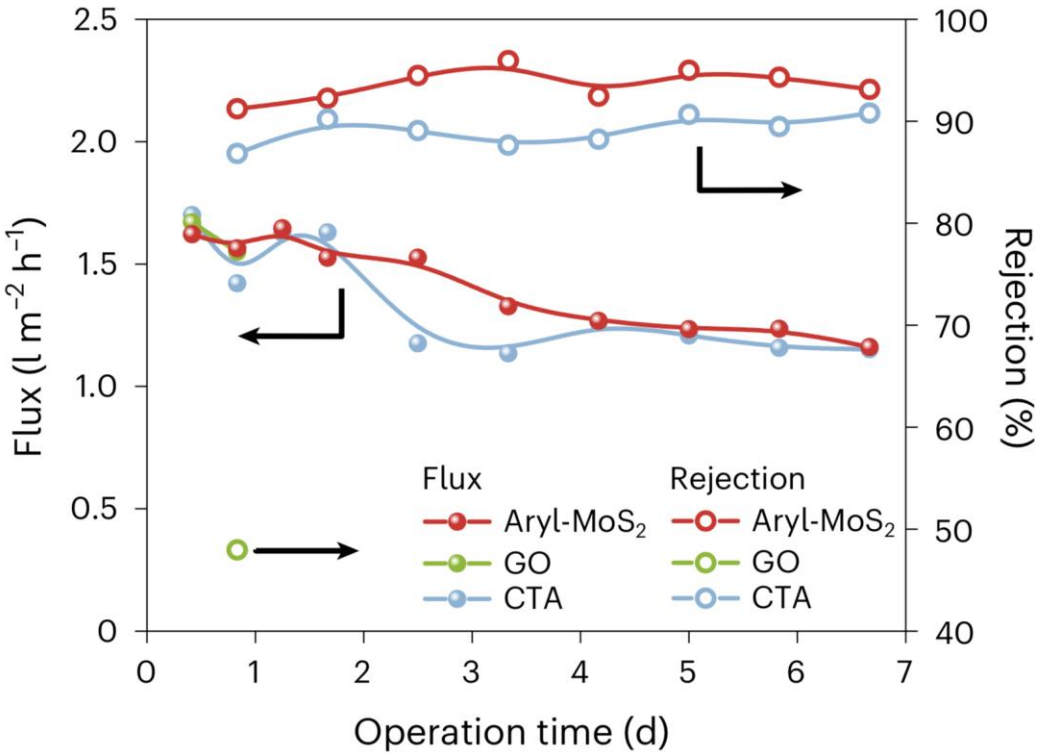


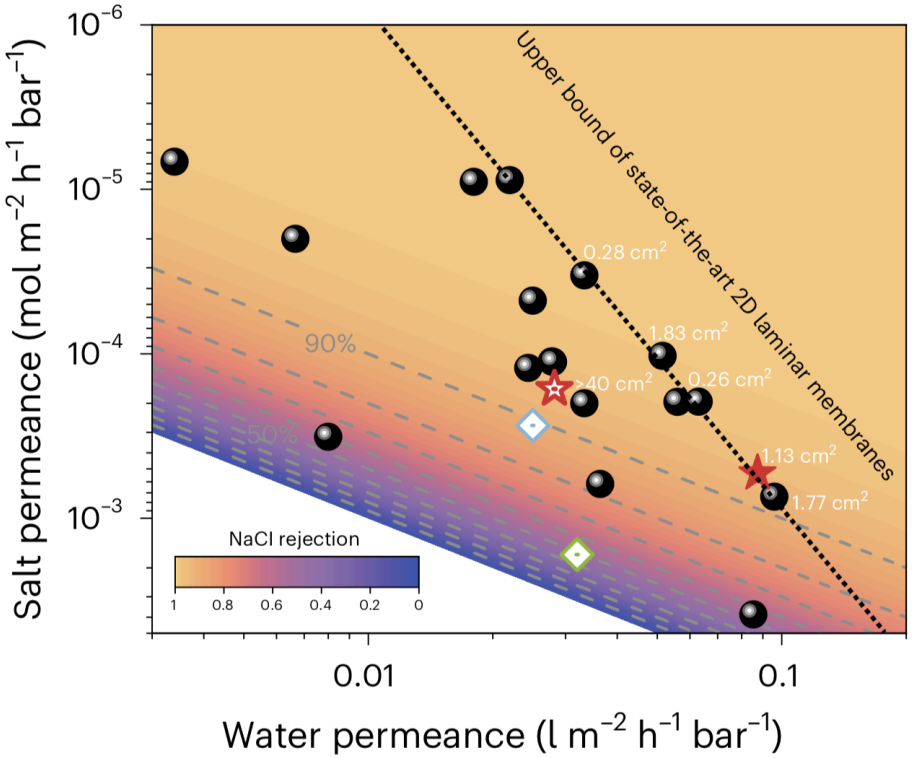
- Finer control of the capillary height
- Importance of stacking disorder on the flux

Wang et al., ***Nature Water***, *2023*

Role of stacking disorders

- Nanolaminate membranes for forward osmosis





- Finer control of the capillary height
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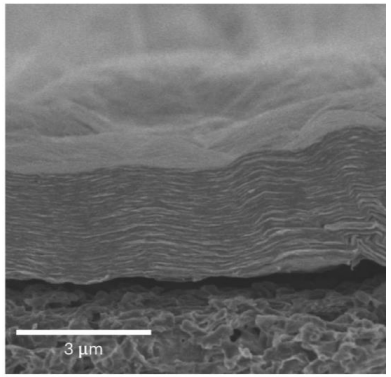
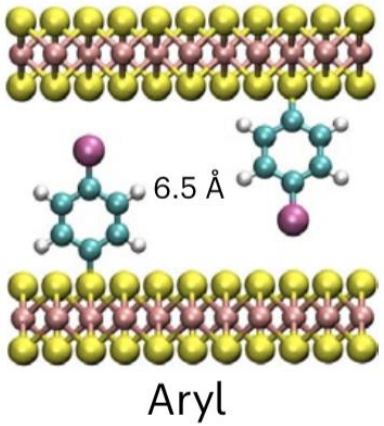
Wang et al., ***Nature Water***, *2023*

Perspectives

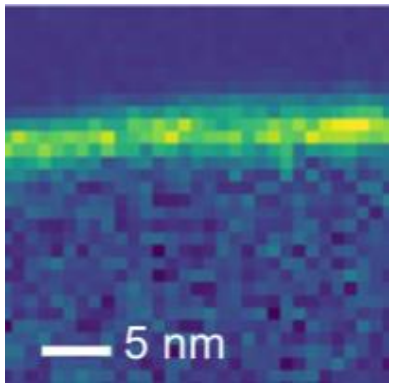
FUNCTIONALIZATION OF 2D MATERIALS

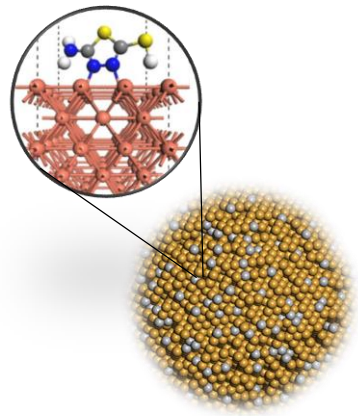




- Control of the surface chemistry
- Control of the interlayer space
- Rejection of NaCl and micro-pollutants
- Molecular weight cutoff: 180 Dalton

MATERIALS ENGINEERING OF COPPER-BASED CATALYSTS





- Rational design of Cu-based catalysts
- Use of electron-withdrawing molecules
- Link between the oxidation state and selectivity for C_{2+}
- Formation of 2-propanol: importance of the alloy quality