One-atom-thick Membrane for Advanced Separation

Vice-Chancellor's Research Day

20th January 2025

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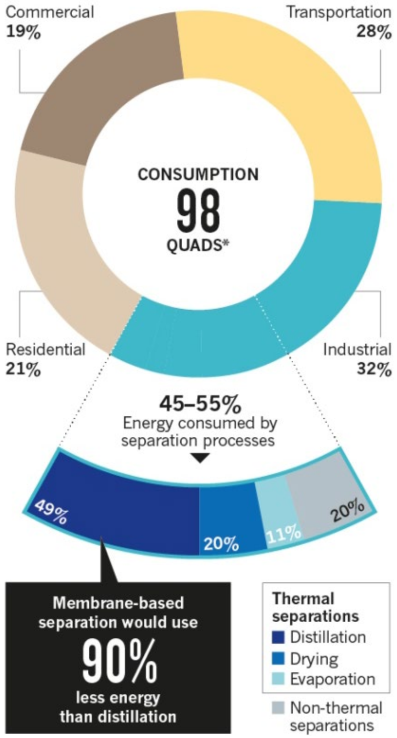
Motivation

Industrial De-carbonization Target



By 2030

- ❑ Switch to 20 TWh of low-carbon fuels (UK)
- ❑ Capture 3 million tons of CO_2 (UK)

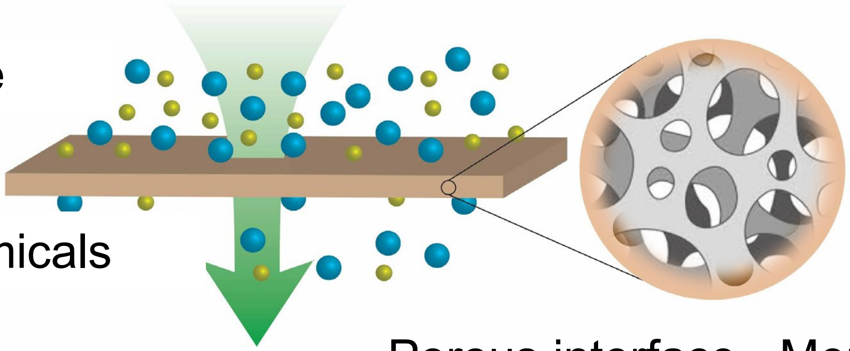
Industrial Decarbonisation Strategy, 2021

P. Lively *et al.* ***Nature*** 532, 435–437, 2016

Energy-Efficient Separation Process

Separation Energy consumption:
6 million tonnes oil/year for UK

Mixture



Pure Chemicals

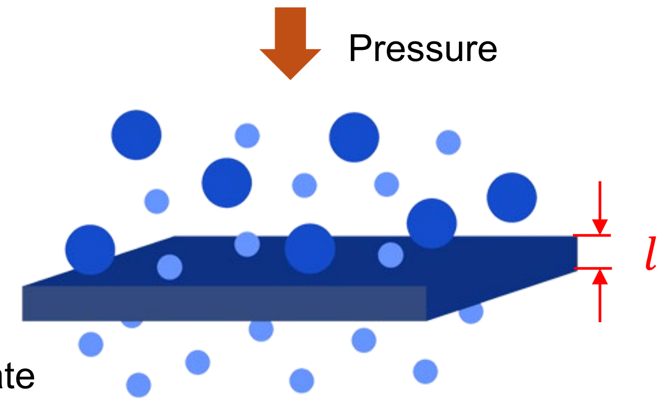
Porous interface - Membrane

Membrane is promising technology to …….

- ❑ Save 90% separation energy
- ❑ Cut down Carbon capture cost < 20$/ton

Energy Consumption in the UK (ECUK) 2023

My Research focus – one-atom-thick membranes

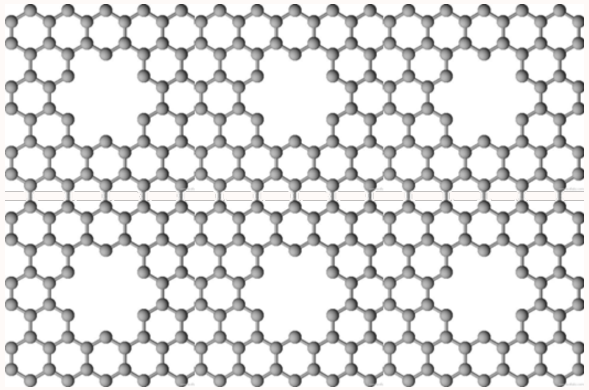


Properties

- ✓ **One-atom-thick layer (e.g. graphene)**
- ✓ **High carrier (electron) mobility**
- ✓ **Unique optical property**

Transform one-atom-thick films as high-performance membranes.

☐ Shortest mass transport channel



High performance membrane

L. Wang *et al.* ***Nature Nanotech*** 12, 509–522 (2017)
L. F. Villalobos, D.J. Babu, K-J Hsu, *et al* ***Acc. Mater. Res.***, 3, 1073-87 (2022)

☐ Unique electronic/optic property

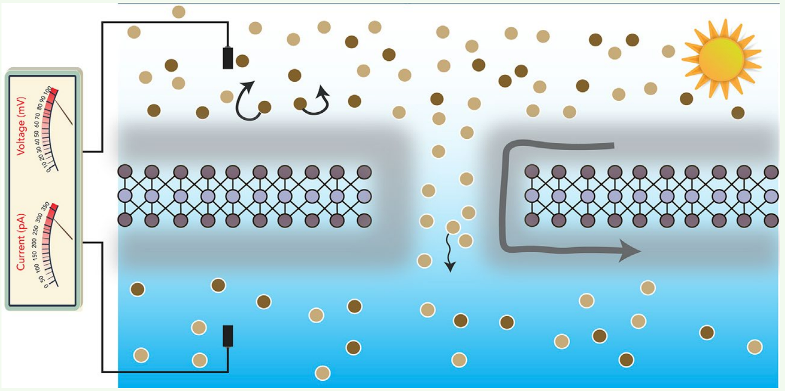


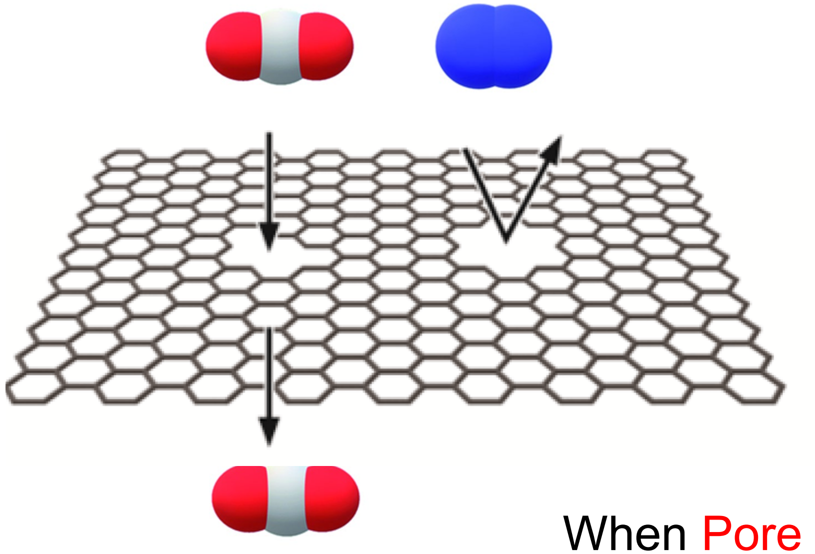
Photo-enhanced mass transport

M. lozada-hidalgo *et al.* ***Nature Nanotech*** 2018, 13, 300–303
M. Graf *et al.* ***Joule*** 2019, 3, 1549–1564

Challenges to incorporate sub-nm nanopores

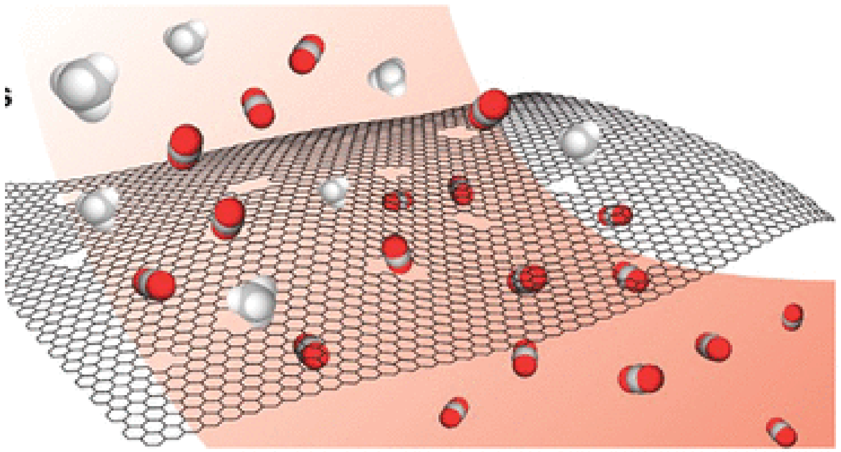
High resolution nanopore

CO_2 (3.3 Å) N_2 (3.6 Å)



1 Å = 10^{-10} m

High density nanopores



When Pore density reach 10^{12} cm^{-2} to 10^{13} cm^{-2}

Expected 10^4 to 10^5 GPU gas permeance

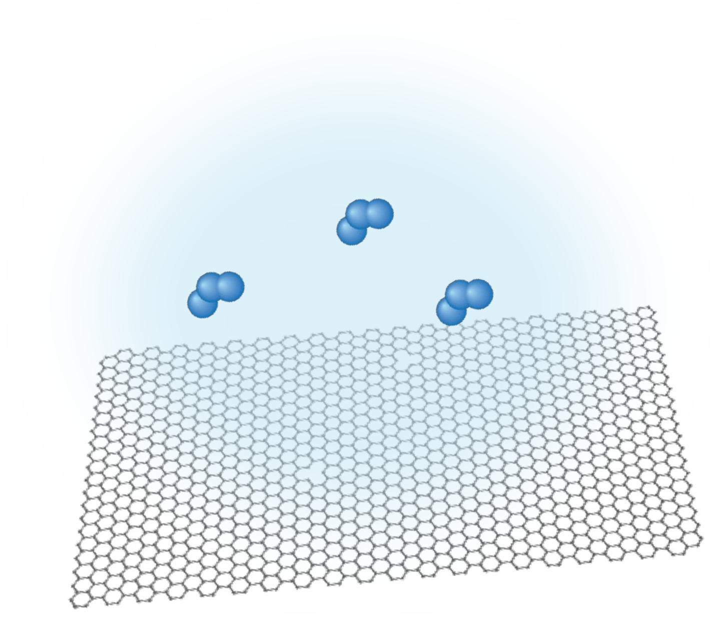
≈ 10 to 100 times of commercial membranes

Yuan *et al.* ***ACS Nano*** 2017, 11, 7974−7987

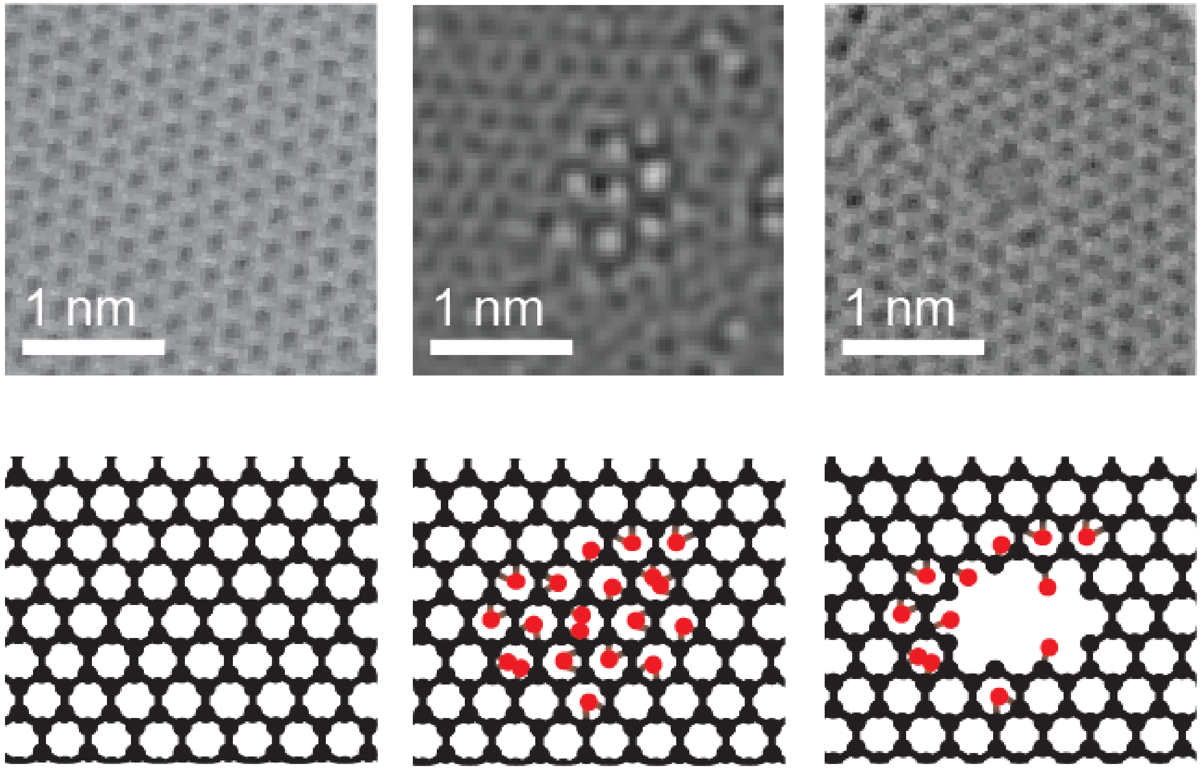
Our approaches

Nanopore engineering → **Mathematical modeling** → **Energy-efficient Membrane**

1. In-situ probe nanopore etching kinetics



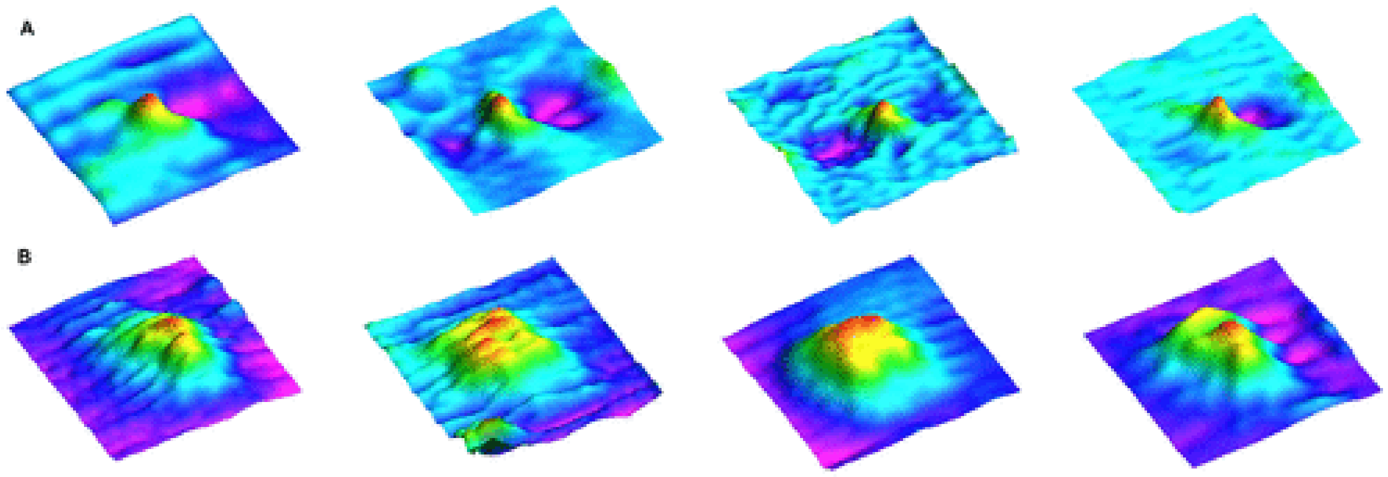
Ozone etching



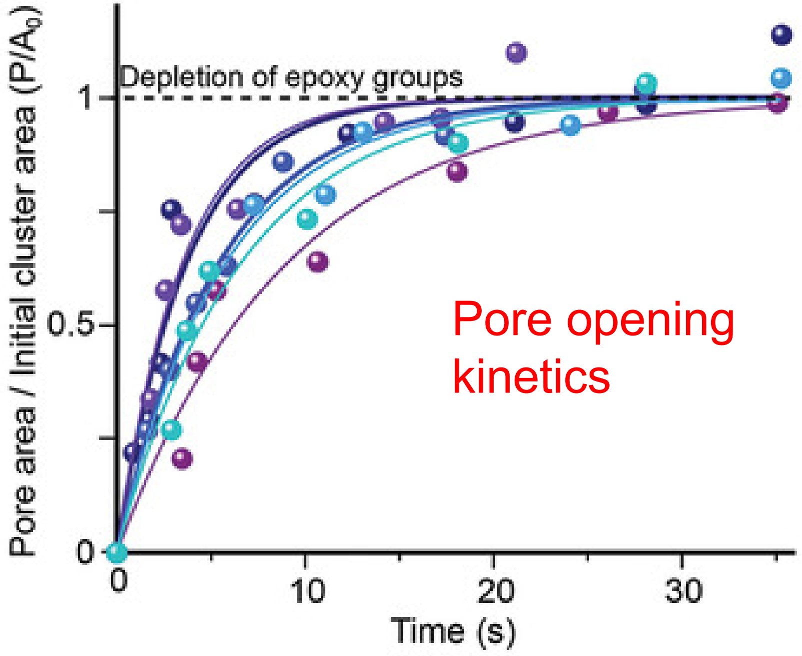
Pristine Graphene | Functionalized Graphene | Nanopore formation

S. Huang, *et al.* ***Advanced Materials***, 2022, 34, 2206627

1. In-situ probe nanopore etching kinetics



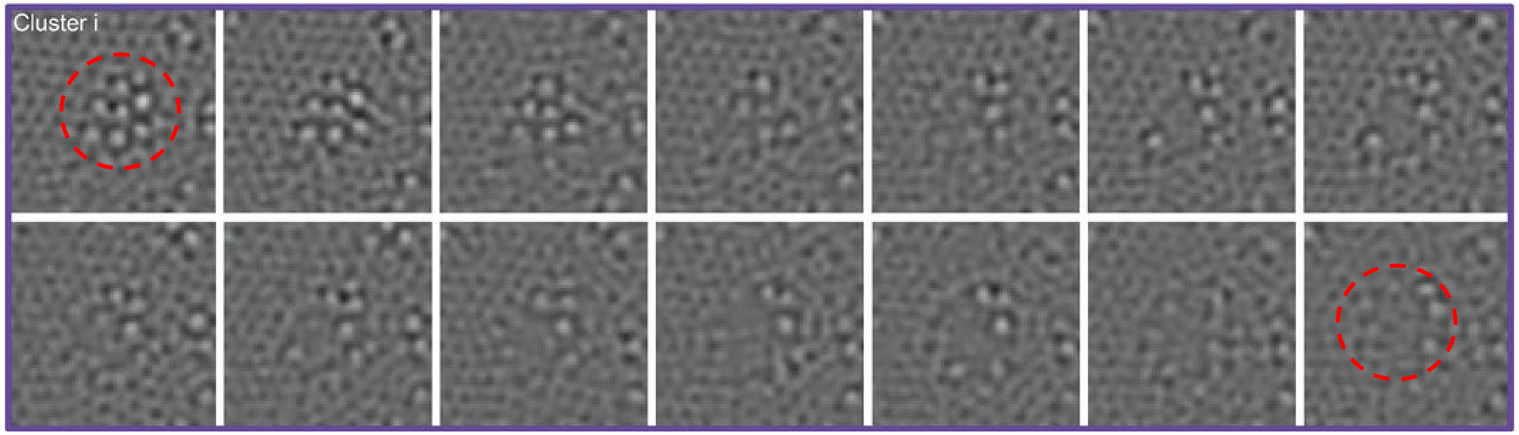
3D STM to illustrate the functional cluster formation



Pore opening kinetics

O-clusters in graphene lattice:
1 h exposure to 10 % O_3 at 43 °C

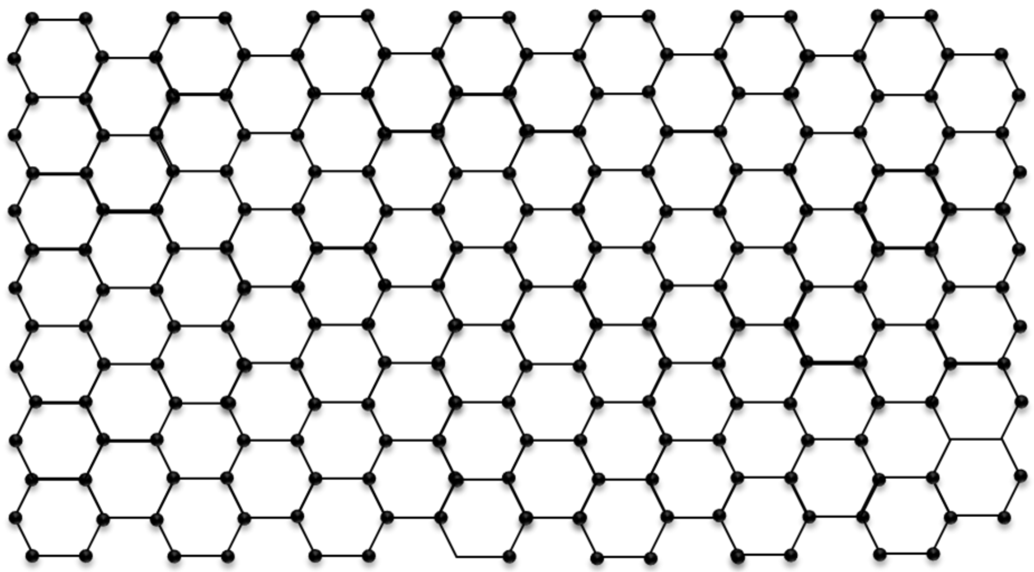
O-cluster to pore:
80-keV beam with a dose rate of 15700 $e^- s^{-1}Å^{-2}$



AC-HRTEM to probe the nanopore opening

S. Huang, *et al.* ***Advanced Materials***, 2022, 34, 2206627
S. Huang, *et al.* ***Science Advances***, 2021, 7(9): eabf0116.

2. Mathematical modelling to guide nanopore formation

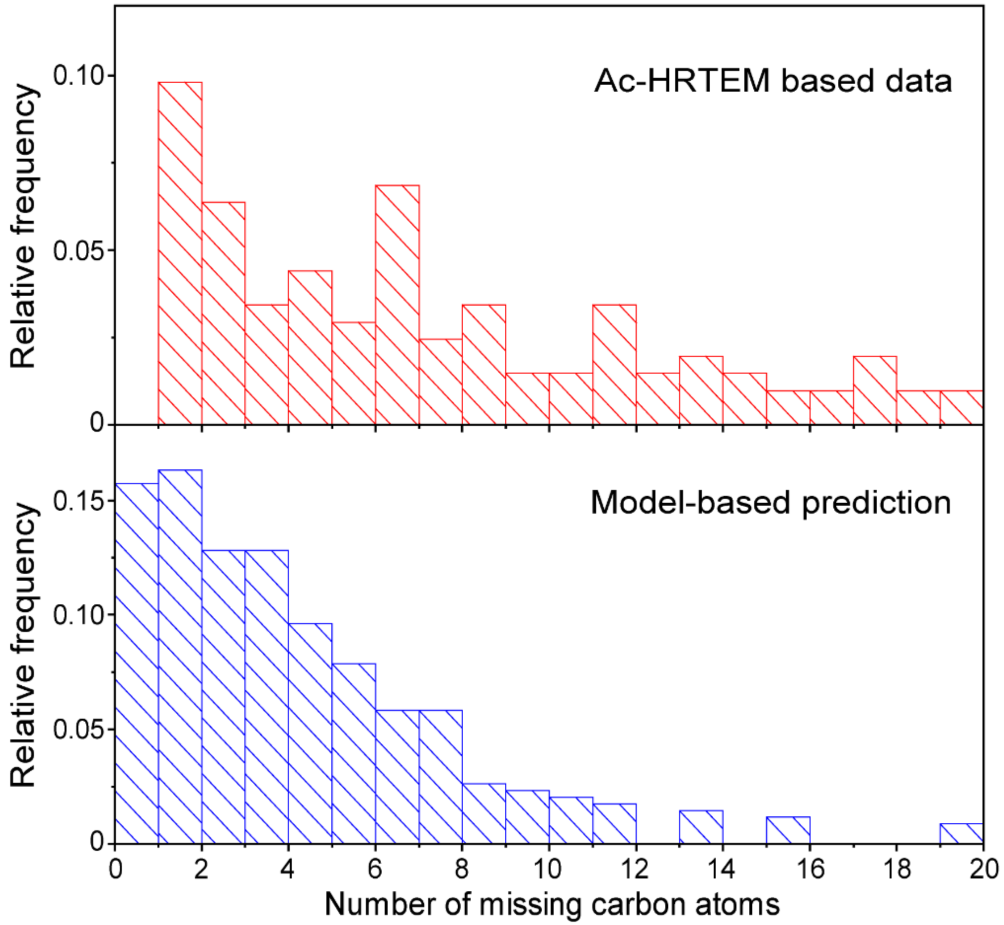


Nucleation sites during [t_{i-1}, t_i]:

$$N_i = C_{\text{density}} k_c H \int_{t_{i-1}}^{t_i} P dt$$

Carbon atom removing of the pores nucleated during [t_{i-1}, t_i].

$$v_i = \sum_{m=i}^{m=n} \frac{\Delta C_m}{\sum_{k=1}^{m} N_k} = \frac{\Delta C_i}{N_1 + N_2 + \ldots N_i} + \ldots\ldots + \frac{\Delta C_n}{N_1 + N_2 + \ldots\ldots + N_n}$$

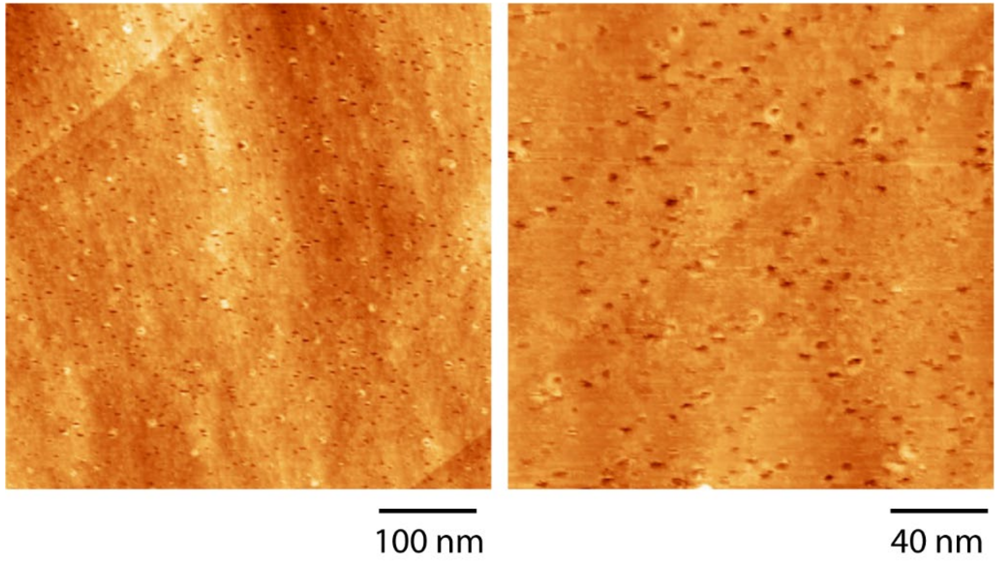


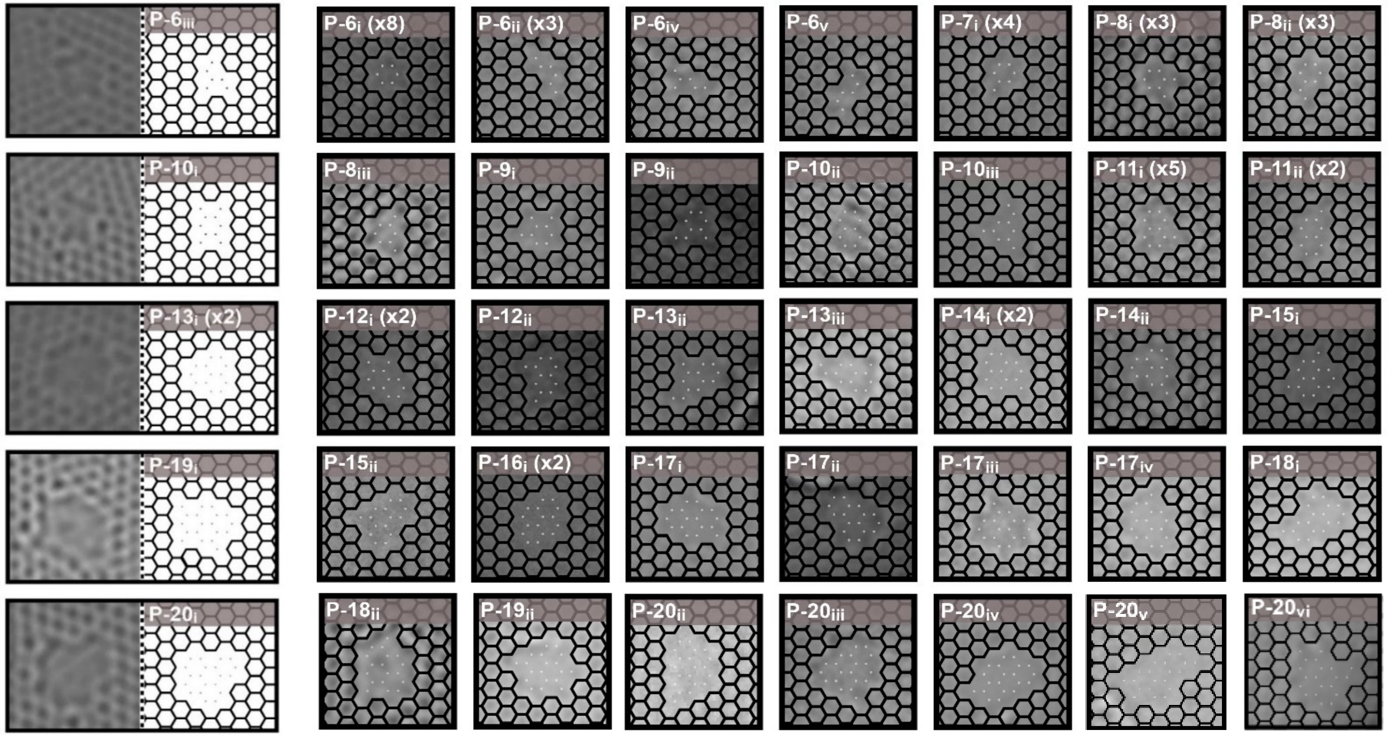
We design our nanopores!

S. Huang *et al.* ***Science Advances***, 2021, 7(9): eabf0116

2. Mathematical modelling to guide nanopore formation

Based on mathematical modelling, optimized experiment condition was chosen to form the following nanopores



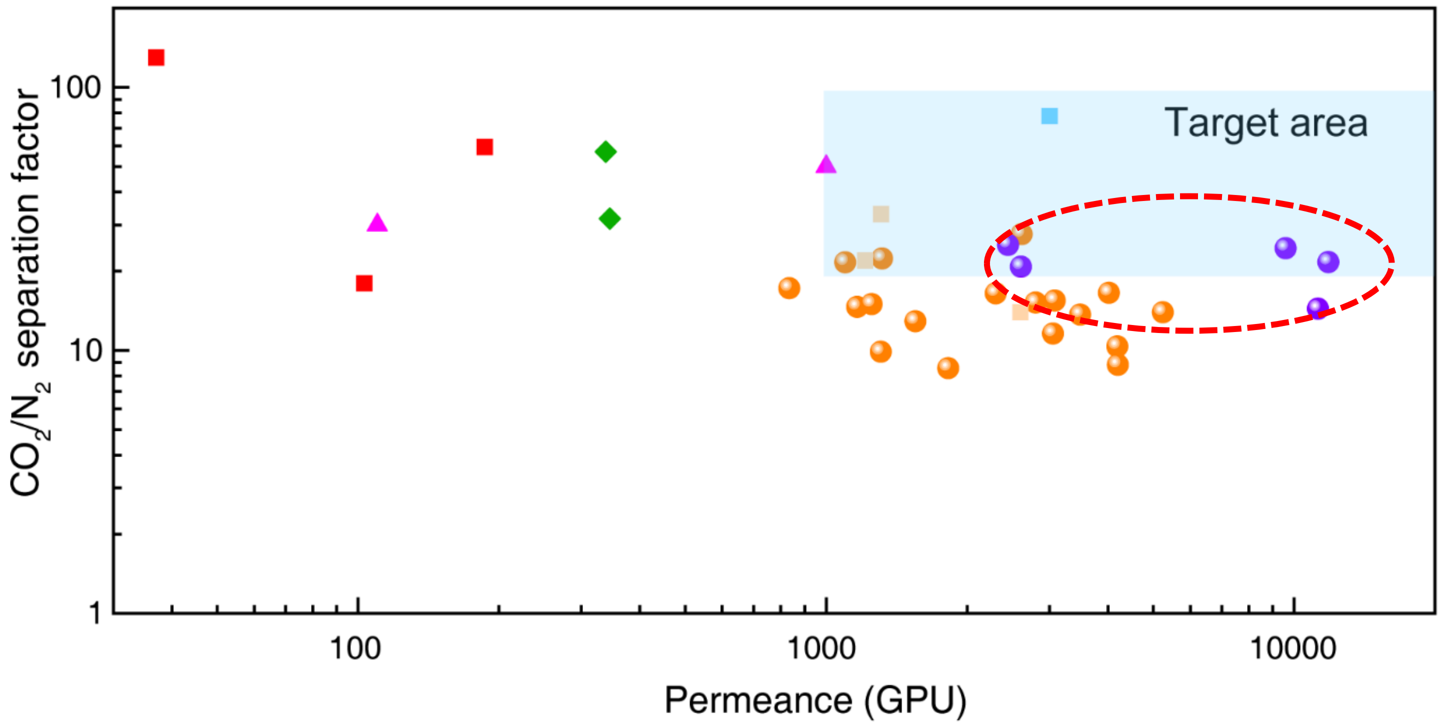


Realize uniform sub-nanometer pore (6 – 20 missing carbon atoms) with high density 1.6 x 10^{12} cm^{-2}

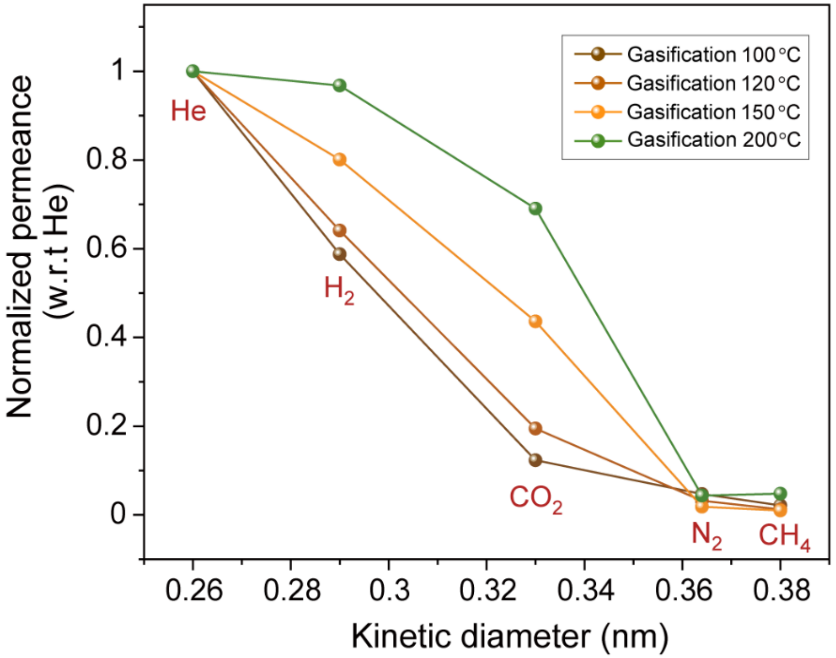
Some of structure observed in experimentally for the first time!

S. Huang, *et al.* ***Advanced Materials***, 2022, 34, 2206627
S. Huang, *et al.* ***Science Advances***, 2021, 7(9): eabf0116.

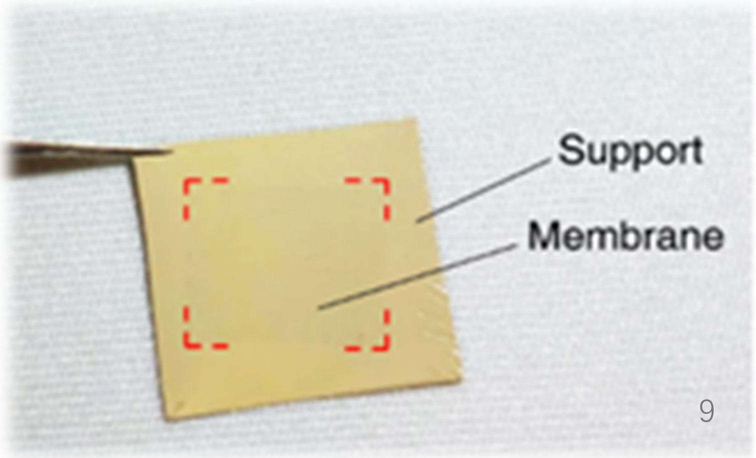
3. The 1st reported one-atom-thick membrane for gas separation



High permeance membrane for CO_2/N_2



Fine tune for different gas pairs



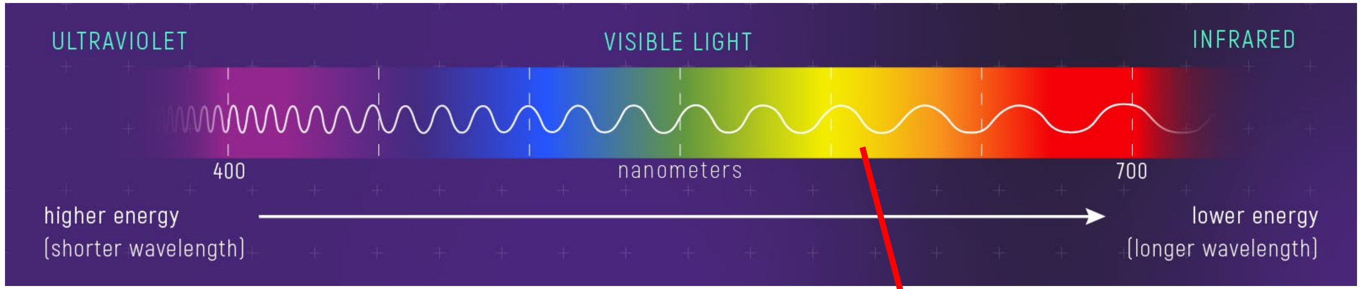
Within 1% porosity
10 times higher permeance than commercial membrane

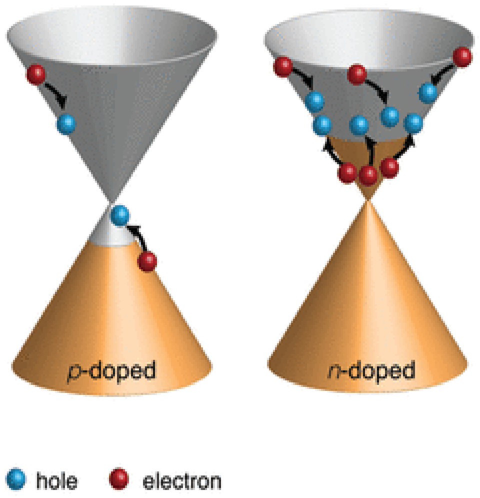
S. Huang, *et al.* ***Advanced Materials***, 2022, 34, 2206627
S. Huang, *et al.* ***Science Advances***, 2021, 7(9): eabf0116.

Can we save more energy?

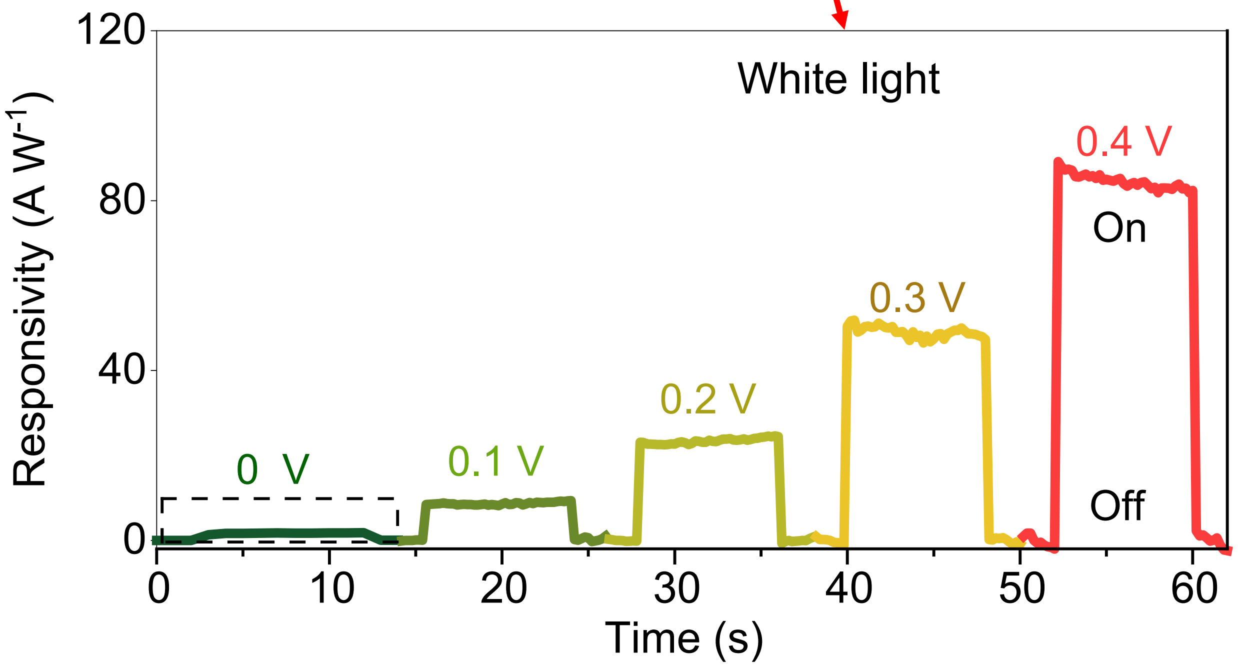
Photo-enhanced graphene membrane

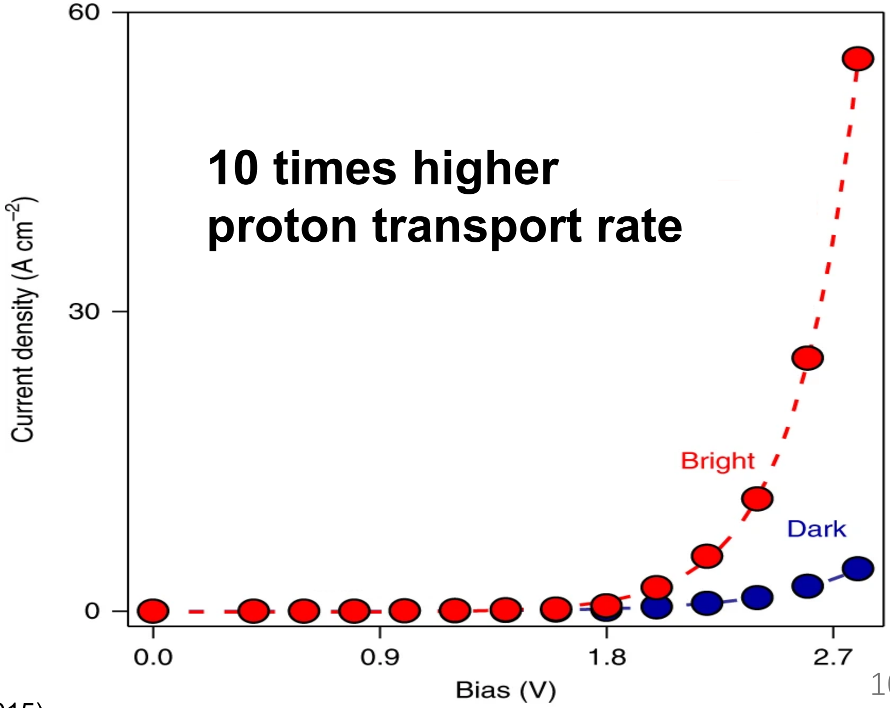
Mass transport ~ Electron structure ~ Photon effect





Photons ~ Electrons



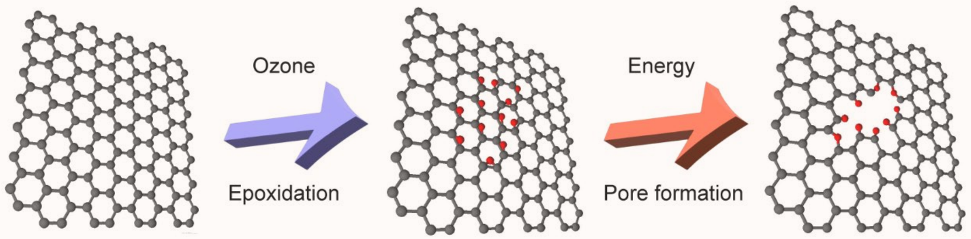


S.Huang et al. ***Nature Commun*** 14, 6932 (2023)
M. lozada-hidalgo et al. ***Nature Nanotech*** 13, 300–303 (2018)
Johannsen JC et al, ***Nano Letters*** 15 326 (2015)

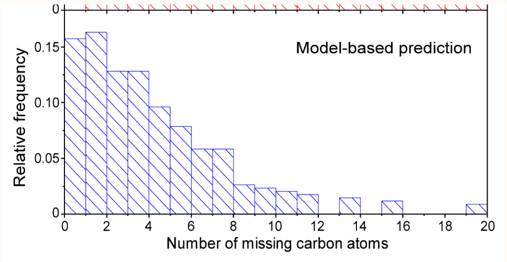
Summary

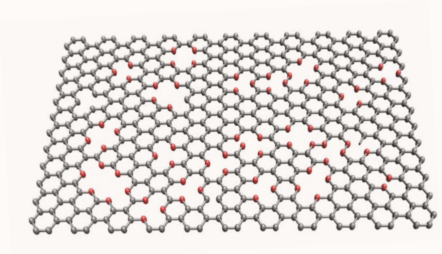
Engineering one-atom-thick graphene as a high-efficiency mass transport interface

Nano Engineering to high performance separation



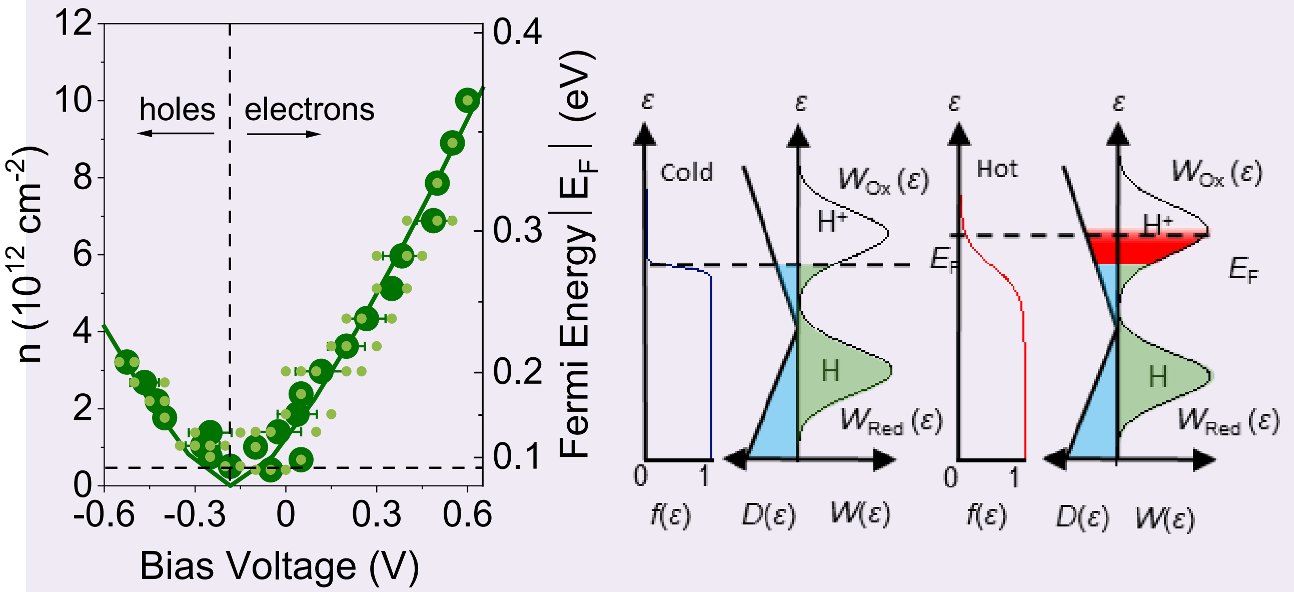
In-situ probe nanopore formation





Mathematical model-guided nanopore fabrication

Unique photo-enhanced phenomenon



Tune Fermi energy control photo-excited electrons

Acknowledgment

Thank you for your attention!