Two-Dimensional Nitrides as Highly Efficient Potential Candidates for CO_2 Capture and Activation

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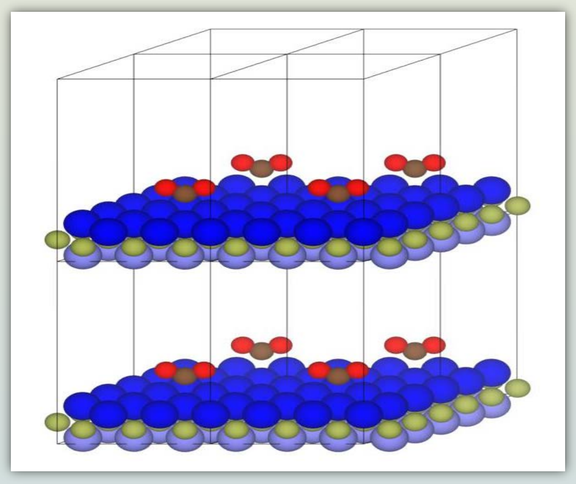
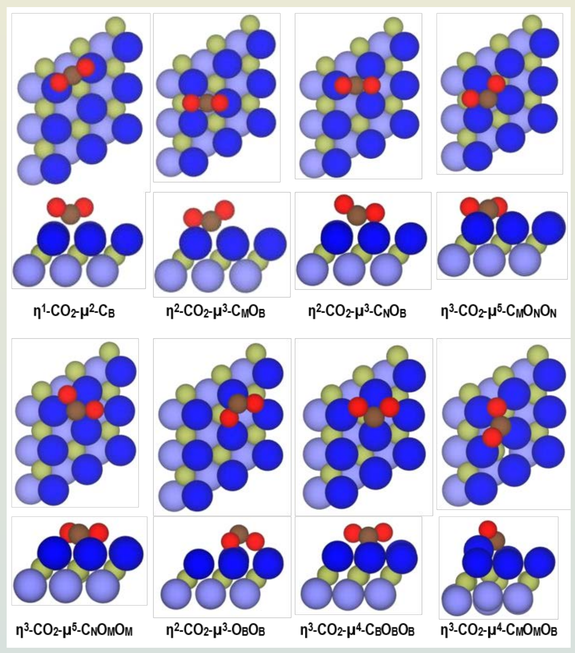
The performance of novel two-dimensional nitride MXene in carbon capture and storage (CCS) is analyzed for a broad range of pressure and temperature conditions. The present theorethical simulations, by using density functional theory (DFT) based calculations, predict that CO_2 can be strongly adsorbed and even activated on the M_2N (M=Ti, Zr, Hf, V, Nb, Ta, Cr, Mo, W).[1]

Computational details:

- (3×3) supercell model
- Code: VASP 5.4.4
- Functional: PBE and PBE-D3
- Cut-off energy of 415 eV.
- Converge criteria: 10^{-5} eV

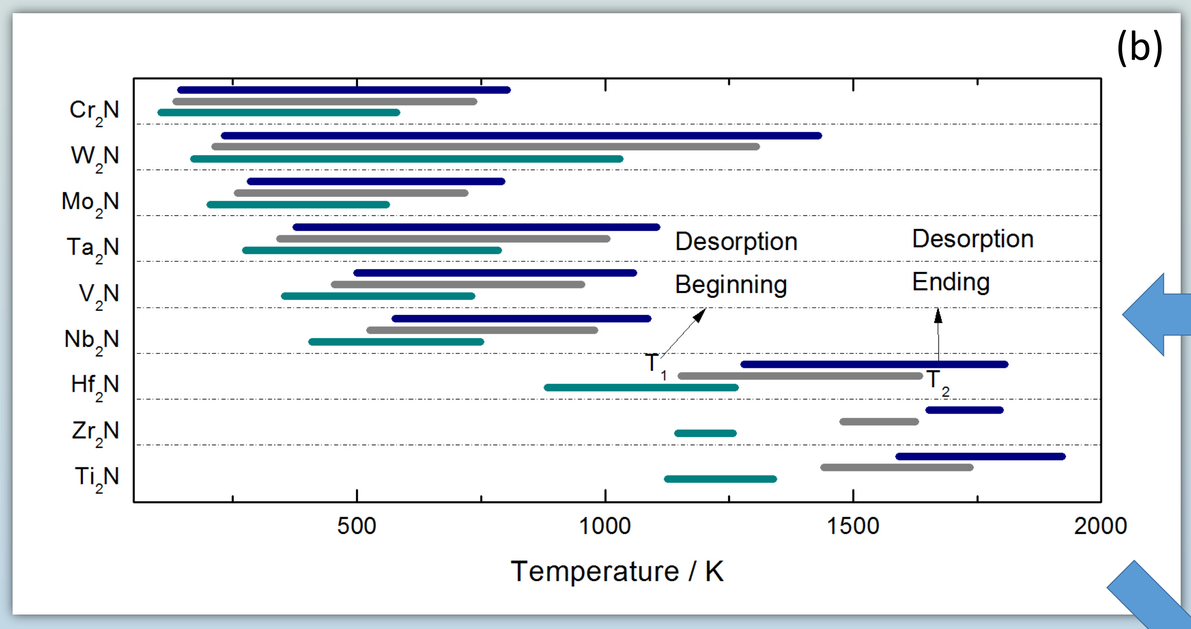


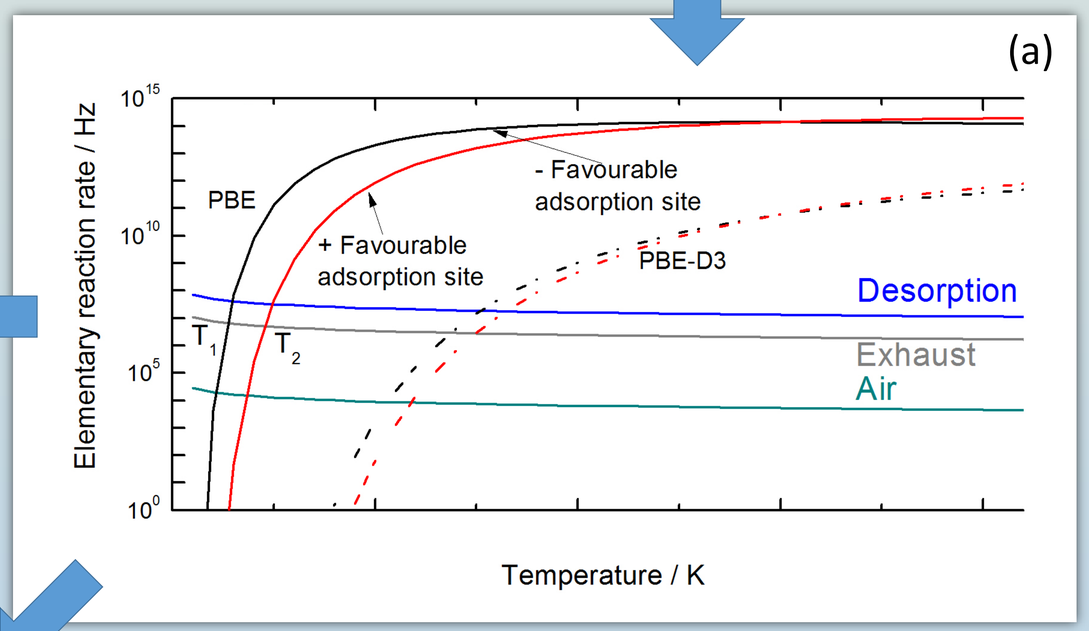


$$E_{ads} = E_{CO_2/slab} - E_{slab} - E_{CO_2} + E_{ZPE}$$

Many adsorption sites have been found for each material with different E_{ads} values.

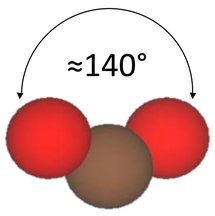
Using the equation of an elementary reaction rate and assuming the adsorption and the desorption are non-activated process, the adsorption/desorption rates can be estimated from the transition state theory (TST) for different temperatures and pressure by using the computed E_{ads} values.[2]

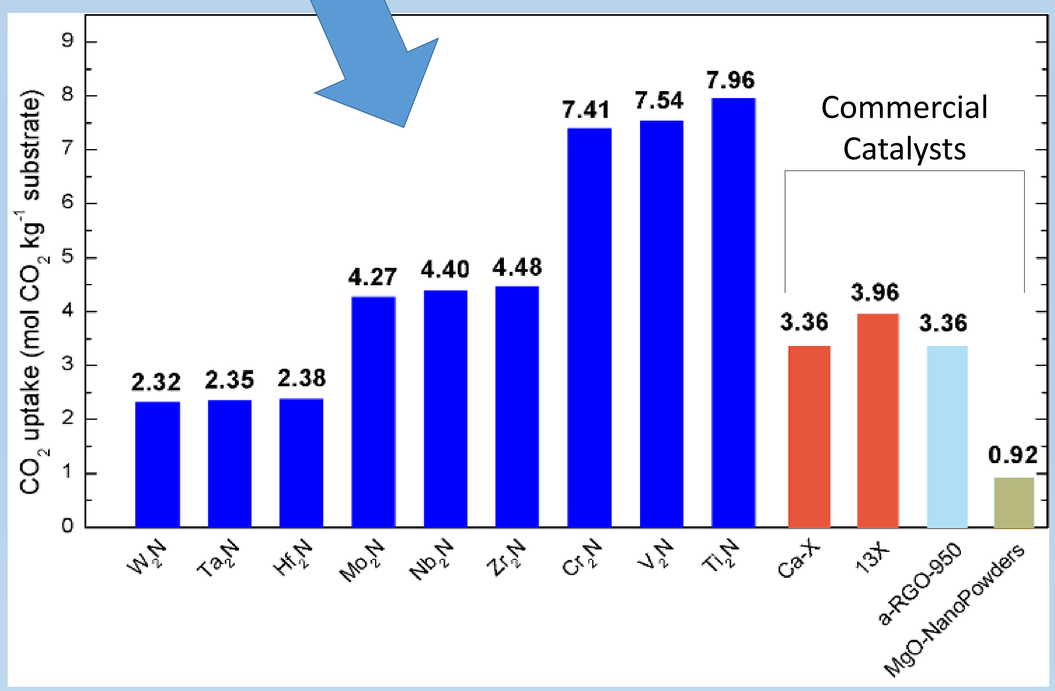




Adsorption/desorption temperature ranges (b), are obtained by the corresponding rates (a) for each material and adsorption site:

- Inside the ranges CO_2 molecule can be adsorbed.
- Wide ranges ⇒ Critical effect of dispersive forces.
- By analizing structural parameters of CO_2 molecule ⇒ Activated!
- There is a huge variety ⇒ Can be they used for different purposes?





Conclusions:

- E_{ads} range between -0.40 to -2.71 eV at PBE level, -1.33 to -3.13 eV at PBE-D3 level. Apparently, dispersive forces do play a key role in the CO_2 adsorption.
- CO_2 molecules are activated when adsorbed in all cases.
- In many MXenes the CO_2 can be released by annealing.

Great capability of CO_2 storage: even in the most conservative conditions, MXene are competitive with nowadays used materials. The usage of nitride Mxenes appears to be more feasible than carbide MXenes.[3]

1. Raul Morales-Salvador, Ángel Morales-García, Francesc Viñes, and Francesc Illas, ***Phys. Chem. Chem. Phys.***, 2018, DOI: 10.1039/C8CP02746C.
2. Christian Kunkel, Francesc Viñes, and Francesc Illas ***Energy Environ. Sci.***, 2016, **9**, 141-144.
3. Ángel Morales-García, Adrián Fernández-Fernández, Francesc Viñes, and Francesc Illas, ***J . Mater. Chem. A***, 2018, **6**, 3381-3385.