

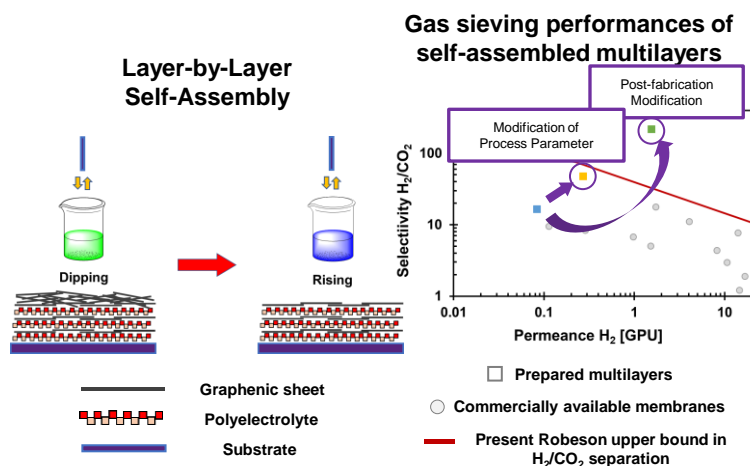
## Graphene multilayers for gas sieving

Giacomo Foli,<sup>a,b</sup> Matteo Minelli,<sup>b</sup> Vincenzo Palermo<sup>a</sup>

<sup>a</sup> ISOF (National Research Council of Italy - CNR), <sup>b</sup> DICAM (University of Bologna - UniBo)

giacomo.foli2@unibo.it

Climate change combined with current geopolitical upsets are seriously demanding a conversion of our energy supply chain. Hydrogen is a carbon-free energy vector that can be prepared from a vast series of resources, even renewable, [1] but a low-cost strategy to isolate it is still challenging. Indeed, independently from the production approach, hydrogen is obtained in gas mixtures, mainly coupled with CO<sub>2</sub>. [2,3] Membrane-based separation can be a suitable solution to prepare pure hydrogen, [4] but perm-selectivity performances of conventional materials are limited by the permeability/selectivity trade-off, named Robeson upper bound. [5] Nanosized composite membranes, fabricated using 2D materials, offer possibility to efficiently size-sieve gas molecules. Graphene-based self-assembled multilayers represent a tunable platform that allows to tailor final molecular architectures and resulting sieving performances, eventually surpassing current Robeson upper bound. Presented multilayers are prepared by an electrostatically ruled Layer-by-Layer self-assembled technique, alternatively exposing a polymeric substrate to a polyelectrolyte solution and a graphenic suspension. Each dipping step is followed by a rinsing in pH=7 water, to remove excess of deposited material (**Figure 1**, left). All perm-selectivities of prepared multilayers are determined by a standard manometric technique. In an attempt to improve poor level of permeability, the properties of the prepared multilayers are modified by post-fabrication modifications or by tuning process parameters of the self-assembly, such as annealing of the self-assembled multilayers or change in the concentration of solution/suspension used for the self-assembly (**Figure 1**, right). All prepared multilayers are extensively characterized in terms of morphology (SEM), elemental composition (XPS) and molecular architecture (XRD).



**Figure 1:** On the left, schematic representation of layer-by-layer self-assembly; on the right, perm-selectivity performances in H<sub>2</sub>/CO<sub>2</sub> separation of some prepared multilayers compared with currently commercially available membranes (present Robeson upper bound in reported, red line).

### References

- [1] S. Singla, *Journal of Environmental Management*, 302 (2022) 113963
- [2] N. Muradov, *International Journal of Hydrogen Energy* 42 (2017) 14058
- [3] S. Albertazzi et al., *Catalysis Today* 106 (2005) 297
- [4] N. Pal, M. Agarwal, *International Journal of Hydrogen Energy*, 53 (2021) 27062
- [5] L.M. Robeson, *Journal of Membrane Science*, 320 (2008) 390