Computational-aided high-throughput screening of hydrogen permeation through graphene-derivative 2D materials: elucidating behaviour for application in proton-conducting membranes

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Abstract

One of the main limitations of proton exchange membrane fuel cells (PEMFC) is the low temperature range in which the membranes can operate. In a collaborative effort in the framework of the RIC2D center, we are working on evaluating different 2D-materials to be used as proton conductive membranes that could operate in conditions where typical polymeric membranes fail (i.e., at 100ºC-400ºC, the infamous 'materials gap' in proton conductors). The work builds on the findings from Sir Andre Geim's group demonstrating that graphene is highly permeable to protons, although completely impermeable to hydrogen molecules or other gases at ambient conditions [1,2].

In addition to graphene, there are hundreds of known two-dimensional materials that have not been explored from a permeability perspective, which is the purpose of this work. To guide the design and selection of promising 2D-structures, Density Functional Theory (DFT) calculations have been used as a high-throughput computational method [3] for the initial screening based on two descriptors related to the permeability of the 2D-membranes: proton transfer barrier and electron clouds. Focus has been paid on the graphene-derivatives (e.g., fluorographene, graphene oxide, graphene, etc.) and their modifications including boron, nitrogen substitutions, among others. Simulations have been validated with experimental results (when available) and used as a guide for the rational selection of the best 2D materials for this application. Furthermore, a machine-learning approach has been developed which removes the computational-time bottleneck [3,4].

References

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Figures

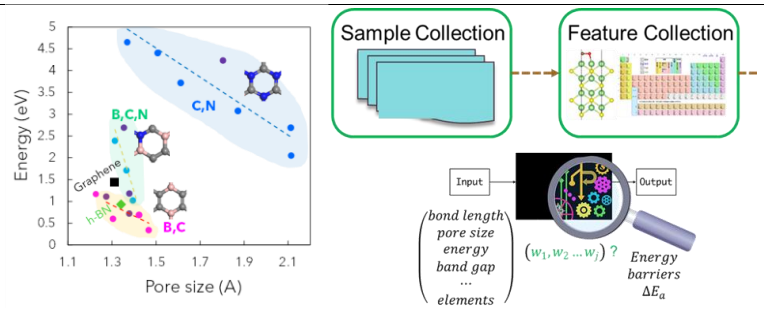

Figure 1: Calculated DFT energy barriers of graphene-derivative structures, and correlation with the relaxed pore size of the permeated ring**; (left)** scheme of machine learning approach used for high-throughput screening of hundreds of 2D materials reported in open-source databases **(right)**.