Quantum-mechanical simulations of hydrogen isotopic separation by graphyne-based membranes

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Deuterium and trititum constitute relevant substances for many scientific and industrial applications. However, they cannot be easily separated from a mixture with H_2, the most abundant hydrogen isotope, as standard methods are based on chemical properties, which are similar for isotopes. For this purpose, in this study [1], we have employed a quantum-mechanical method that takes into account mass-dependent effects. Graphdiyne, a recently synthesized porous derivative of graphene [2], offers a promising opportunity to enhance these effects, as its subnanometric pores enable a confinement of the hydrogen molecules when travelling through them. In our simulations the molecules have been treated as pseudoatoms and their interaction with the membrane is represented by an improved Lennard-Jones force field [3]. By performing three-dimensional time-dependent wavepacket propagations [4], we have obtained the transmission probability of each isotope, which in turn has been used to compute the permeances at different temperatures. Finally, we have calculated the selectivity of deuterium and tritium over hydrogen (shown in Figure 1) as the quotient of their permeances: It is seen that the selectivities (favoring the heavier isotopes) increase as temperature decreases until reaching a maximum value. The maximum is a result of a compromise between zero point energy and tunnelling effects, which benefit the heavier and lighter species, respectively. These results are promising for efficient deuterium and tritium separation from hydrogen.

References

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Figures

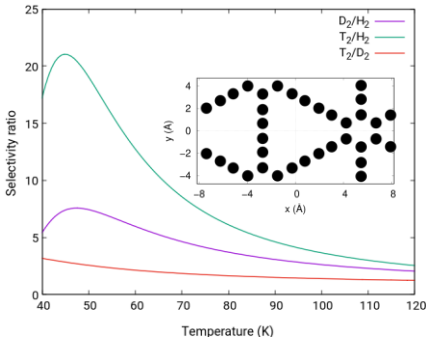

Figure 1: Selectivity as a function of temperature, inset shows the graphdiyne unit cell