Assessing the permeability of monolayer graphene oxide to protons and gases

Lourdes F. Vega[1,2]

Daniel Bahamon[1,2], Maryam Khaleel[1,2], Marcelo Lozada-Hidalgo[1,3,4], Andre K. Geim[3,4]

[1] *Research and Innovation Center for graphene and 2D materials (RIC2D), Khalifa University, PO Box 127788, Abu Dhabi, United Arab Emirates*

[2] *Research and Innovation Center on CO_2 and Hydrogen (RICH Center) and Chemical Engineering Department, Khalifa University, PO Box 127788, Abu Dhabi, United Arab Emirates*

[3] *Department of Physics and Astronomy, The University of Manchester, Manchester, UK*

[4] *National Graphene Institute, The University of Manchester, Manchester, M13 9PL, UK*

Lourdes.vega@ku.ac.ae

Two-dimensional materials (e.g., graphene and h-BN) offer a prospect of membranes that combine negligible permeability to gases with high proton conductivity, and could outperform the polymer-based existing proton exchange membranes used in various applications including fuel cells [1,2].

Functionalized graphene structures such as graphene oxide (GO) and fluorographene have attracted considerable interest as a proton conductor because of its high lateral proton conductivity along the basal plane. This in-plane conductivity makes them a promising additive to Nafion and fosters interest in proton-conductive multilayer films [3]. However, little is known about proton and molecular transport across the basal plane of such monolayers.

Here we show that relatively large, micrometer-scale areas of pinholes-free functionalized monolayers (i.e., of GO) are impermeable to gases but exhibit proton conductivity through the basal plane which is nearly two orders of magnitude higher than that of graphene and boron nitride [1,4]. The results have been complemented with quantum and molecular simulations to elucidate the physical insight into the phenomena occurring during the proton permeation. In particular, the enhancement is attributed to microscopic corrugations of the underlying graphene lattice, which are caused by functional groups bonded to the graphene surface.

These findings demonstrate that chemical functionalization of 2D crystals can be utilized to enhance their proton transparency without compromising gas impermeability, which could expand their potential applications in hydrogen-related technologies.

References

[1] S. Hu, et al. Nature 516 (2014) 227-230.

[2] P.Z. Sun, et al. Nature 579 (2020) 229–232.

[3] T. Bayer, et al. J. Membr. Sci. 541 (2017) 347–357.

[4] Z.F. Wu, P.Z. Sun, O.J. Wahab, et al. Nat Commun 14 (2023) 7756.

Figures

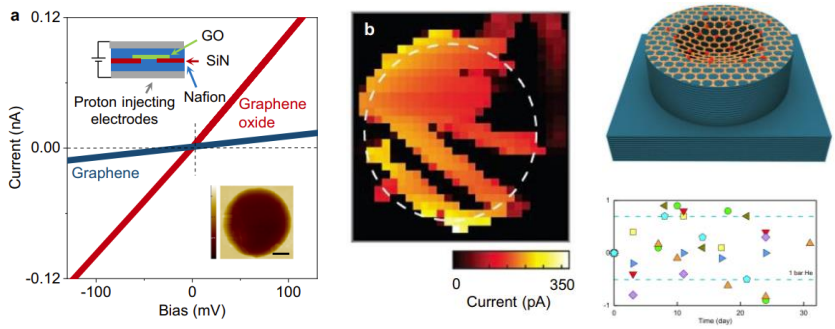

Figure 1: a) I-V characteristics for graphene and GO monolayers. [Top inset: schematic of the devices used in the measurements]. **b)** Example of scanning electrochemical cell microscopy (SECCM) maps for monolayer GO. **c)** Schematic of GO microcontainer devices, and changes in sealing height for Helium gas with time (different symbols correspond to different devices).