Field effect control of proton transport and hydrogenation in double-gated graphene

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Graphene's basal plane can function as a perfectly selective barrier permeable to protons but impermeable to all ions and gases, stimulating its use in applications such as membranes, catalysis and isotope separation. Protons can also chemically adsorb on graphene and hydrogenate it, inducing a conductor-insulator transition intensely explored in graphene electronic devices. However, both processes face energy barriers that in the case of proton transport motivate strategies to accelerate it, such as introducing vacancies, incorporating catalytic metals or chemically functionalising the lattice, but these can compromise other properties like ion selectivity or mechanical stability. Here we show that independent control of the electric field E~ V nm^{-1} and charge carrier density n~10^{14} cm^{-2} in double gated graphene allows decoupling proton transport from lattice hydrogenation and can accelerate proton transport such that it approaches the limiting electrolyte current in our devices. Proton transport and hydrogenation can be driven selectively with precision and robustness that enables proton-based logic-and-memory graphene devices with orders-of-magnitude on-off ratios. Our results show that field effects can accelerate and decouple electrochemical processes in double-gated 2D crystals and demonstrate the possibility of mapping such processes as a function of E and n – a fundamentally different technique to study 2D electrode-electrolyte interfaces.