

## Alejandro García-Miranda Ferrari

Dale A. C. Brownson and Craig E. Banks

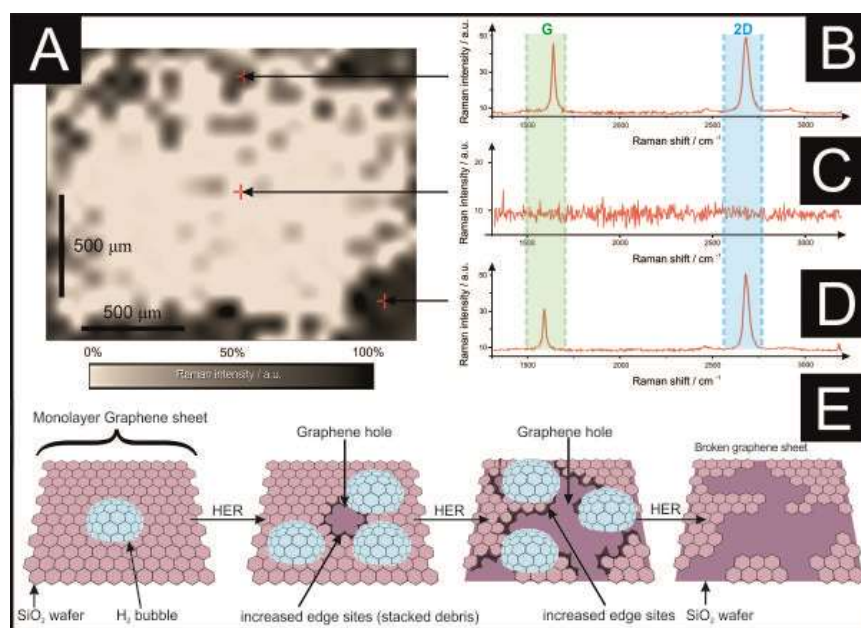
Manchester Fuel Cell Innovation Centre, Manchester Metropolitan University, Manchester M1 5GD, UK.

alejandro.garcia-miranda-ferrari@stu.mmu.ac.uk

# Investigating the Integrity of Graphene towards the Electrochemical Hydrogen Evolution Reaction (HER)

Mono-, few-, and multilayer graphene is explored towards the electrochemical Hydrogen Evolution Reaction (HER). Careful physicochemical characterisation is undertaken during electrochemical implementation revealing that the integrity of graphene is structurally compromised. Electrochemical perturbation, in the form of electrochemical potential scanning, as induced when exploring the HER using monolayer graphene, creates defects upon the basal plane surface that increases the coverage of edge plane sites/defects resulting in an increase in the electrochemical reversibility of the HER process. This process of improved HER performance occurs up to a threshold, where substantial break-up of the basal sheet occurs, after which the electrochemical response decreases; this is due to the destruction of the sheet integrity and lack of electrical wiring/conductive pathways. Importantly, the severity of these changes is structurally dependent on the graphene variant utilised. This work indicates that multilayer graphene has more potential as basis of an electrochemical platform for the HER, rather than that of mono- and few-layer graphene. There is huge potential for this knowledge to be usefully exploited within the energy sector and beyond.

## Figures



**Figure 1:** (A) Monolayer graphene following 20 LSV scans. Part (B) depicts the Raman profile near a hole, showing that it is few-layer graphene, with the characteristic ratio of the G/2D peaks near to 1:1. (C) Depicts the Raman profile of a broken area where there is no characteristic graphene peak (or signal) present. (D) Shows the Raman profile of an intact area where there is monolayer graphene including its typical G (1590 cm<sup>-1</sup>) and 2D (2690 cm<sup>-1</sup>) peaks. (E) Is a schematic representation of the behaviour identified within this figure (A–D), where the emergence of a bubble on the graphene surface (due to the HER) leads to the creation of some rips when the bubbles explode/move. The debris created due to the graphene breakdown is stacked in areas near to the holes/rip. When many bubbles explode, there is an incremental rise of the edge sites caused by the broken graphene pieces, which eventually lead to the complete destruction of the graphene sheet.