356g Catalytic Role of Defective Carbon Substrates in the Dissociation of Water

Milen K. Kostov, Erik E. Santiso, Aaron M. George, Marco B. Nardelli, and Keith E. Gubbins

A necessary step towards the achievement of a hydrogen-based economy is the development of a production process that is able to drastically revamp the energetic cost while leaving, at the same time, a smaller environmental footprint than the current industry standards. Chemical reactions are often carried out in nano-structured media, where the reaction mechanism can be dramatically changed due to the interactions of reacting species with the substrate. One point of interest is the recent experimental evidence for stable defects in graphene layers such as vacancies. Here we propose an alternative route for the production of hydrogen, where the catalytic role of defective carbon substrates is exploited for improving the efficiency of the thermal splitting of water at low temperatures. Using state of the art first principles modeling techniques, we have explored the potential of defective sites in carbon nanotubes to lower the activation energy barrier of dissociation reactions for small molecules. We found that water can dissociate following many possible reaction pathways, some of which have activation barriers lower than half the value for the dissociation of bulk water. These novel reactions enhance the hydrogen yield and the reaction rate suggesting a process of hydrogen production with great scientific and economical implications. Finally, we will discuss this exploration in the context of a complete cycle of energy storage and release through the production of hydrogen in defective carbon substrates.