A COMPUTATIONAL AND EXPERIMENTAL APPROACH TOWARDS THE KNOWLEDGE OF CATALYTIC SITES FOR OXYGEN REDUCTION REDUCTION IN N-DOPED CARBON MATERIALS

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Abstract

The design of advanced N-doped carbon materials as catalysts for oxygen reduction reaction requires a detailed knowledge of the nature of the active sites. There is an important piece of research seeking to overcome this challenge through experimental or computational studies. However, studies that combine both of them are scarce and are necessary to deepen into the knowledge of this key issue.

This work, that combines sophisticated computational calculations and experimental results measured with N-doped carbon materials, provides clear evidence about the nature of the active sites in N- and N- O- containing carbon materials. All carbon materials containing N- and N- O- species have been obtained from polyaniline by heat treatment in different atmospheres (both oxygen-containing and inert atmospheres) and different temperatures (from 600-1100ºC). The computational calculations have been obtained from DFT simulations. This work evidences that edge-type quaternary nitrogen is a highly efficient active site towards oxygen reduction reaction through the four electrons pathway. Nevertheless, this work also demonstrates that other nitrogen species, such as pyridinic or pyrrolic nitrogen, for example, do not produce a higher activity than a non-doped carbon material. On the other hand, oxygen atoms near the nitrogen functionalities modify the chemical nature of the active sites, what could be highly beneficial towards the oxygen reduction catalysis.