Título: ADVANCED AMPEROMETRIC NANOCOMPOSITE SENSORS BASED ON CARBON NANOTUBES AND GRAPHENE: CHARACTERIZATION, OPTIMIZATION, FUNCTIONALIZATION AND APPLICATIONS

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Resumen: Among the wide range of nanocomposites, the incorporation of conducting nanostructured carbon materials, such as carbon nanotubes (CNTs) and graphene, into an insulating polymeric matrix is a very attractive way to combine the unique mechanical and electrical properties of individual filler with the advantages of polymers. Concretely, carbon¿based nanocomposite materials have played a leading role in the analytical electrochemistry field, particularly in (bio)sensor devices, due to their interesting advantages regarding to a pure conductive material, such as versatility, durability, easy surface regeneration, facile incorporation of a variety of (bio)modifiers or low background current, among others.

Accordingly, this Thesis tackles the development of advanced amperometric nanocomposite sensors that after having been optimized regarding to carbon/polymer composition ratios, can be tunable with different types of nanoparticles (NPs) for improving their electroanalytical response.

The electrical properties of these nanocomposites and, therefore, their analytical applicability, are directly influenced by the conducting particles nature and the amount and spatial distribution of them through the insulating polymeric matrix. One of the most important electrochemical properties of these materials is the

similarity of their electrochemical behavior with a microelectrode array. Thus, an optimization of the carbon/polymer ratio with respect to the nature of the conducting material will allow to achieve a better dispersion of the conducting areas through the non-conducting areas, presenting similar benefits to the microelectrode array. In addition, it is known that some parameters, such as composite resistivity, heterogeneous electron transfer rate, material robustness and background capacitance current are strongly influenced by the physical nature of the raw CNT sample, such as their diameter/length ratio and purity, fact that may strongly influences the final electroanalytical response of the transducer material.

Under this context, the first step of this Thesis consisted of implementing a group of instrumental techniques that, systematically applied, have allowed the characterization and optimization of nanocomposite materials composition based on CNTs and epoxy resin (Epotek H77) in relation to the nature of the raw CNT sample for the fabrication of improved electrochemical sensors. The developed characterization protocol includes electrical, electrochemical, morphological, microscopic, spectroscopic and electroanalytical tools.

Having been optimized the CNT/epoxy composition ratios, the next step consisted of enhancing the analytical performance of these electrochemical nanocomposite sensors introducing an electrocatalytical effect by the incorporation of different NPs. For this goal, a simple methodology for synthesizing a wide range of different NPs has been developed. Intermatrix Synthesis (IMS) has been used as a green technique to design three different alternative routes for CNT/epoxy nanocomposite electrodes modification, which offer a customized way for the preparation of more sensitive amperometric sensors.

Finally, the characterization and functionalization studies applied for CNT¿based electrochemical nanocomposite sensors have been extended for nanocomposite materials based on another allotropic form of carbon: the graphene, which is the novel application in terms of nanostructured carbon material.