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FROM NANO- TO MACRO-SCALE: HYBRID MAGNETIC CARBON NANOCOMPOSITES AS A TOOL FOR CATALYTIC WET PEROXIDE OXIDATION

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Introduction

There is a need for technological innovation in the water sector worldwide, as a result of the growing demand for water supplies – meeting increasingly restricted quality criteria, while coping with the increasing scarcity of clean water sources[1]. Under this context, the use of treated wastewater as an alternative water source has emerged as a topic of high priority[2]. However, meeting current quality requirements for wastewater reuse is a great challenge, in which nanotechnology holds a great potential[1].
Catalytic wet peroxide oxidation (CWPO) is a promising water/wastewater treatment technology; it enables the formation of highly oxidizing hydroxyl radicals ($HO^{\bullet}$) under atmospheric pressure and low to moderate temperatures, when a suitable catalyst is employed for the decomposition of hydrogen peroxide (H_2O_2)[3]. However, further improvement of catalyst design is still required in order to allow the scale-up of the CWPO technology towards real-scale applications.
Bearing this in mind, our work has been focused on the development of highly active and stable hybrid magnetic carbon nanocomposites for CWPO. A detailed catalyst design at the nanoscale, based on the understanding of the surface reactions and interactions involved in the CWPO process, has recently allowed us to move forward towards the treatment of a real industrial wastewater with high pollutant load – collected from a mechanical biological treatment (MBT) plant for municipal solid waste processing. This communication reports the findings obtained in the last four years by our research group in this quest. A particular emphasis is given to the synergistic effects arising from the combination of iron-based catalysts with the easily tuned properties of carbon-based materials.

Results and Discussion

A hybrid magnetic graphitic nanocomposite – composed by a magnetite core and a graphitic shell, was synthesized by hierarchical co-assembly of magnetite nanoparticles and carbon precursors. Following its application in CWPO, it was concluded about the role of the carbon shell. Specifically, the encapsulation of magnetite nanoparticles within carbon frameworks (i) enhances the catalytic activity in CWPO when compared to bare magnetite, while (ii) strongly limiting the leaching of iron species to the treated water. The first effect was ascribed to the increased adsorptive interactions between the carbon phase and the pollutant molecules, which enable a more intimate contact between the generated $HO^{\bullet}$ radicals and the pollutant. The lower metal leaching obtained with the nanocomposite catalyst was ascribed to the confinement effect promoted by the carbon shell. As a result of these positive effects, very high pollutant mass removals were obtained with a rather high efficiency of H_2O_2 consumption, and the composite catalyst was active for operating pH in the range 3 – 6[4].

Seeking for catalyst optimization, the individual effects of the metal species employed in the nanostructured composites have been studied. It was found that the simultaneous incorporation of cobalt and iron into carbon matrices (iii) increases the rate of HO$^{\bullet}$ radicals formation, and (iv) promotes a more efficient reduction of Fe^{3+} to Fe^{2+}, thus limiting iron leaching[5].

Based on the findings reported above, it was possible to design a high performance hybrid magnetic graphitic nanocomposite ($CoFe_2O_4$/MGNC) – composed by a cobalt ferrite core and a graphitic shell. In this way, the positive effects described in (i) to (iv) were combined in the same nanocomposite material. It was found that the application of this new generation catalyst enables the treatment of waste waters with high pollutant loads, such as that from the MBT plant considered in this study (cf. **Figure 1**). The biodegradability of the wastewater was enhanced during the treatment performed at pH 6, regardless of its high organic and inorganic content; disinfection was achieved and the treated water revealed no toxicity against selected bacteria. The high stability of $CoFe_2O_4$/MGNC for CWPO was demonstrated in a series of five CWPO reaction/magnetic separation sequential experiments in the same vessel[6].

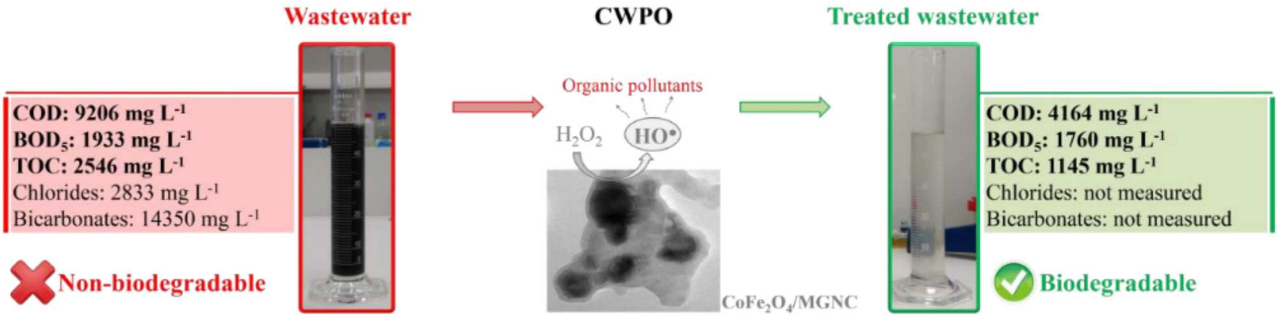

Figure 1. Representation of the CWPO treatment performed in the presence of $CoFe_2O_4$/MGNC.

Conclusions

The proper understanding of the catalyst properties at the nanoscale is fundamental in order to design materials with potential to be an effective tool for real-scale CWPO applications.

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