## Abstract

The present PhD thesis entitled "Functional silica materials for controlled release, sensing and elimination of target molecules" is centred in the design and development of organic-inorganic hybrid materials through the application of supramolecular chemistry concepts. Several materials based on silica have been prepared and characterized for different applications and were presented on this thesis.

The first part of the thesis is centred on the development of materials based on silica being able to modify their fluorescent behaviour depending on the presence or absence of a target analyte on the media. These materials are based on silica nanoparticles acting as inorganic scaffolding, the surface of which is functionalized with two different units: a coordination and a signalling unit (a fluorophore molecule). The interaction between the target analyte (in this case, anions) and the coordination unit will modulate the fluorescent emission properties of the fluorophore. Thus, different materials have been prepared wherein the fluorescent moiety is rhodamine whereas the coordination moiety is imidazolate or a guanidinium salt, respectively. Once both materials have been characterized, their behaviour upon different anionic species at different concentrations was studied, these materials being selective to benzoate (imidazolate functionalized material), dihydrogen phosphate and hydrogen sulphate (guanidinium functionalized material).

The third chapter of the thesis is focused on the application of organic-inorganic hybrid materials for the sensing and elimination of highly toxic species such as neurotoxic agents. These are organophosphorous compounds able to affect seriously the central nerve system. On a first approximation, the molecular gate concept is used for sensing neurotoxic agents. To do that, an inorganic mesoporous silica scaffold is used (MCM-41), whose pores are loaded with a dye acting as signalling molecule whereas the external surface of said scaffolding is functionalized with a molecule being able to react with those neurotoxic agents. That molecule forms a hydrogen bonding network that blocks the pore outlets. In the presence of DCP (diethylchlorophosphate, a neurotoxic agent mimics), and after it has reacted with that molecule, a spatial rearrangement takes place, thus allowing the release of the dye. This way, the presence of neurotoxic agents is signalized with a colour change. In a second approximation, the use of inorganic supports such as MCM-41 for the elimination of neurotoxic agents is studied. To do that, this siliceous material was treated with different bases. As a consequence of this basic treatment, the silanols on the silica surface are deprotonated originating the corresponding silanolates (strong nucleofiles). These silanolates are able to react with the neurotoxic agents yielding to their decomposition and thus, enhancing their elimination from a polluted medium.

Finally, the use of organic-inorganic hybrid materials being functionalized with molecular gates for controlled release applications was studied. The hybrid materials consists on a mesoporous scaffolding based on silica whose pores are loaded with a cytotoxic compound (camptothecin) and its external surface is functionalized with a gluconamide derivative. The presence of a dense monolayer of gluconamides in the outer surface of the material inhibits the release of the cytotoxic compound. Upon addition of enzymes being able to hydrolyse amide bonds (amidase and pronase) the release of camptothecin takes place. The behaviour of the material was proved *in vitro* and *in vivo* using HeLa cells.