# Design and development of novel layered nanostructured hybrid materials for environmental, medical, energy and catalytic applications

Potsi, Georgia

# Summary

The field of layered nanostructured hybrid materials focuses on the synthesis and possible applications of materials that combine the properties of building blocks of nanometer size dimensions. Bringing together the building blocks can enhance their properties or generate new properties not present in either of the constituents alone.

The aim of this thesis centred on layered nanostructured hybrid materials for environmental, medical, energy and catalytic applications, is to describe the synthesis, characterization and possible applications of a number of hybrid layered materials based on carbon allotropes or different inorganic matrices like clay minerals.

Chapter 3 presents the synthesis of multi-functional pillared layered materials synthesized by the intercalation of adamantylamine into the interlayer space of graphite oxide and layered aluminosilicate nanoclays. Different characterization techniques demonstrated the successful intercalation of adamantylamine and showed that the final pillared materials have an increased specific surface area. In addition these hybrids were found to be capable of adsorbing significant quantities of organic pollutants, which entails a great potential for environmental remediation applications. Moreover they were found to present improved cytotoxic activity on A549 cancer cells, whilst the cytotoxicity towards MRC-5 cells (normal) was minimal, a fact that renders them suitable as antiproliferative agents in biomedical applications.

Chapter 4 describes the chemical oxidation of carbon nanodiscs, industrially prepared via the so-called pyrolytic Kværner Carbon Black & $H_2$ process, towards the formation of a hydrophilic analogue. The detailed

characterization of the pristine and the oxidized material is described and the study of the cytotoxic properties of the oxidized nanodiscs is reported. Besides resulting in the separation of carbon nanodiscs from the mixed nanodiscs/nanocones/soot starting material, the oxidation treatment causes the attachment of oxygen-containing functional groups (epoxy, hydroxyl and carboxyl groups) on the nanodisc surface, improving the solubility in polar solvents and thereby the use in various applications. The study of the cytotoxicity properties showed that the oxidized nanodiscs can act as cytotoxic agent and promises well for their future use in nanobiocatalytic systems.

Chapter 5 presents a review of various experimental studies of the synthesis and properties of carbon nanostructures containing organic-inorganic cage-like polyhedral oligomeric silsesquioxane (POSS) nanoparticles The aim is to illustrate the improvements of chemical and physical properties that can be achieved by the combination of POSS with different carbon nanostructures focusing on the potential impact of these hybrid nanostructures on various technological applications.

Chapter 6 reports the intercalation of iron substituted ($Fe^{+3}$) cubic silsesquioxanes in a sodium and an acid-activated montmorillonite to form novel catalytic pillared structures. A variety of characterization techniques was applied to prove the successful intercalation of the cubic silsesquioxanes into the clay matrices as well as the formation of pillared structures after calcination. The final pillared hybrids possess high specific area and contain α-$Fe_2O_3$ (hematite) nanoparticles as verified by Mössbauer spectroscopy. Catalytic measurements showed that the final hybrid pillared materials catalyse the conversion of isopropanol to diisopropylether and propene due to high specific area and presence of acid sites on the surface; the selectivity is affected by stereochemical parameters.

Chapter 7 presents the fabrication of metal ($Cu^{2+}$ and $Fe^{+3}$) decorated POSS thin films via the Langmuir –Schaefer method or via a combination of this method with self-assembly, using a simple surfactant such as arachidic acid (AA). Characterization with different techniques proofed the successful deposition of the layers, leading to a periodically repeated AA-Metal ($Cu^{2+}$ and $Fe^{+3}$)-POSS-AA unit. Moreover the interlayer distance between the units was found to be affected by the coordination of the metal ions. Additionally, comparison of the two fabrication protocols showed that the hybrid films deposited following synthetic route involving self-assembly process lead to better ordered structures.