TOP-DOWN AND BOTTOM-UP APPROACHES FOR THE SYNTHESIS OF FULLERENE-BASED PILLARED LAYERED STRUCTURES

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Abstract
Due to their outstanding physical and chemical properties, low-dimensional assemblies, where order and organization follow supramolecular principles, have assumed significant importance the last decades. Controlling both the organization of the assemblies and their physical and chemical properties through simple external parameters the creation of new tailored functional materials could be achieved. Towards this aim, the insertion and the subsequent behavior of pure C60 and a series of fullerene derivatives (fulleropyrrolidines, fullerols and polybromo-fullerenes) into the interlayer space of layered materials using top-down and bottom-up synthetic approaches will be presented. Smectite clays, graphene oxide and graphite nitrate were used as layered host materials exploiting their templating properties. Top-down methods include simple intercalation reactions in bulk where neutral fullerene derivatives and pure C60 were inserted into either organically modified or not layered matrices, while water-soluble and positively charged fullerene derivatives were introduced into the aluminosilicate clay galleries through ion exchange. The nature of the microenvironment between the 2D nanometer size sheets regulates the topology of the intercalated molecules enabling molecular alignment in a two-dimensional assembly. Moreover, a new bottom-up approach for the production of hybrid materials where layered materials act as the structure directing interface and reaction media will be also described. This new method, based on combining self-assembly with the Langmuir Schaefer technique, uses the layered nanosheets as a template for the grafting of fullerene molecules in a bi-dimensional array, and allows for perfect layer-by-layer growth with control at the molecular level. The resulted fullerene-based pillared layered structures were characterized by a combination of analytical techniques. The experiments gave an insight into the formation process, structural details and properties of the final pillared structures. The reported composite materials constitute new hybrid systems where C60 differs from its crystals or its solutions while the described fabrication routes open new perspectives for the design and construction of two-dimensional functional hybrid materials.

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