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Controlling molecular and nanoscale structures and compositions of porous functional silica hybrid materials: role of interactions at interfaces

CORINE GERARDIN

University of Montpellier

Presenting Author: corine.gerardin@enscm.fr

Ordered mesoporous silica resulting from the synergy between supramolecular self-assembly of macromolecular compounds and sol-gel processes present unique porous textural and surface properties, which make them attractive as drug delivery systems, sensors, adsorbents, catalysts, ion conductors and more generally for various applications in the fields of health, environment and energy.

The control of their structure at the nanoscale is mainly governed by the interactions between the amphiphilic entities and the silica precursors under specific physicochemical conditions of the synthesis. Different types of interactions (hydrogen bonding, van der Waals, electrostatic complexation) occur simultaneously and their cooperativity or competitiveness determine the structural parameters of the obtained materials : system curvature at the nanoscale, pore size, nature of the mesophase and wall thickness.The functionalization of their mesopores is also a key step in their preparation for applications. Functionalization by organic, organometallic or bioactive groups can be achieved by well-controlled routes. But the introduction of polymers in the mesopores remains a major challenge. Yet, this could confer new properties to the materials. Conventional routes for polymer post-functionalization of mesopores do not allow to obtain dense and homogeneous distributions of polymers at the nanoscale. The use of polyion complex micelles (PICs) as structuring agents can overcome the problems encountered. PIC micelles are dynamic assemblies obtained by electrostatic complexation between a double-hydrophilic block copolymer and a polyelectrolyte auxiliary of micellization. Their formation is reversible in water as a function of pH and ionic strength. Their use is of major interest since they allow (1) the simple control of mesostructures (2D hexagonal, lamellar, 3D spherical cubic) and pore sizes, (2) the recovery and recycling of pore-forming polymers, and (3) the direct preparation of structures functionalized by homogeneously distributed polymers. Another great advantage of the use of PIC micelles is the easy preparation of mesoporous nanoparticles of controlled size. Finally, the study of their formation mechanisms showed that the nature of the polymer blocks and their preferential interactions with silica, the micellization auxiliary or the solvent proved to be key parameters in controlling not only the internal porous interface of the material, but also its external surface.