Polymer and Colloid Highlights

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Low-temperature Preparation of Functional Carbon Nanocapsules *via* Self-assembly and Carbonization of Hexayne Amphiphiles

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Carbon nanostructures with a defined number of extended dimensions constitute promising components for high performance composites, lithium storage materials, or nanoelectronics.^[1] Mitigating their poor dispersibility in organic matrices by chemical functionalization should allow for the preparation of novel types of electronically active nanocomposites. The typical methods to prepare carbon materials, however, involve high-energy processes that impede the tailored preparation of carbon nanostructures with a controlled morphology and chemical functionalization.^[2] In this context, we developed a novel strategy for the low-temperature wet-chemical preparation of carbon nanocapsules based on the synthesis, self-assembly, and subsequent mild carbonization of oligoyne amphiphiles as reactive molecular precursors.[3] A novel synthetic protocol based on the Negishi reaction was crucial to couple two sp-hybridized carbon atoms with high yields so that we obtained the desired molecular precursors from simple starting materials on the multigram scale.^[4] In this way, we prepared glycosylated hexaynes that resembled typical glycolipid amphiphiles. Our investigations showed that these carbon-rich amphiphiles reversibly self-assembled in aqueous solution to form bilayer vesicles the size of which was controlled by vesicle extrusion (Fig. 1). In the aggregated state, the reactive hexayne segments of our amphiphiles were densely packed, and UVirradiation at 1 °C sufficed as a mild external stimulus to induce the carbonization process. After 12-16 h of UV irradiation, the hexayne segments were completely consumed according to absorption spectroscopy, and characterization of the obtained carbon nanocapsules provided us with conclusive evidence for an extensive carbonization. The carbon microstructure was analyzed by Raman spectroscopy; we found that the nanocapsules' walls were constituted of graphite-like amorphous carbon, which usually requires process temperatures above 600 °C. At the same time, the carbohydrate shell was retained as verified by a glucose binding assay with Concanavalin A. Thus, the carbononly reactive oligovne segments of our amphiphiles allowed us to prepare functional carbon nanocapsules under very mild conditions. Our current efforts focus on demonstrating the universal nature of this approach as a means to obtain functional carbon nanosheets, nanotubes, as well as carbon micropatterns.

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Fig. 1. (Left) Schematic representation of the wet-chemical preparation of carbon nanocapsules from glycosyl functionalized amphiphiles containing a reactive carbon-only hexayne segment. (Middle) Unilamellar vesicles were obtained after self-assembly of the carbon-rich amphiphiles in water and vesicle extrusion through polycarbonate membranes. (Right) The carbonization of these vesicles then proceeded under UV-irradiation below room temperature and the formed carbon nanocapsules comprised a graphite-like amorphous carbon with a hydrophilic carbohydrate shell.