SYNTHESIS AND CATALYTIC APPLICATIONS OF DENDRIMER-CARBON NANOTUBES HYBRIDS

Antonin Desmecht, Sophie Hermans, and Olivier Riant

Institute of Condensed Matter and Nanosciences, UCLouvain
Place Louis Pasteur 1, 1348 Louvain-la-Neuve, Belgium

For the past decades since their discovery, nanocarbons (NCs) such as carbon nanotubes (CNTs) and graphene (G) have drawn the attention of the scientific community for their outstanding properties. Indeed, these materials exhibit an exceptional resistance to mechanical and chemical stress, combined with excellent electrical and thermal conductivities and high surface area. Moreover, bundles of CNT and G form a mesoporous network, hence preventing any diffusion limitations when suspended in a liquid. These properties in addition to the dedicated efforts of both academic and industrial groups have enabled the emergence of various applications involving NCs, such as sensors, energy storage and optoelectronic devices, medicinal tools and catalytic materials. Since the first report of catalytic application in 1994, the interest in developing catalysts based on NC is swiftly raising, paralleling their decreasing mass-production costs. Moreover, the peculiar properties of NCs can be put at use in catalytic applications hence NCs distinguish themselves from other more conventional heterogeneous supports.

In order to immobilize an organometallic complex at NC's surface, the latter needs to be chemically altered. This is typically achieved through covalent or non-covalent functionalization.[1] With the final goal of catalytic application in mind, we decided to investigate first the two-step nucleophilic addition of organolithium compounds / electrophile capture for NC functionalization. Herein we propose the use of unprecedented electrophiles to react with NC reduced by *n*-BuLi: propargyl bromide[2] and isopropoxyboronic acid pinacol ester.[3] In addition, we combined the exceptional properties of NCs with those of polyamidoamines (PAMAM) dendrimers.[4] Indeed, PAMAM dendrimers allow increasing the number of surface functions since one grafting point on NC's surface can lead up to dozens of terminations depending on the dendrimer's generation. The dendrimer's structure itself can also act as an encapsulating host for metallic nanoparticles.

We synthesized homogeneous supported iridium complexes at the extremities of the dendrimers for imine reduction application. In addition, our materials were used as template for the encapsulation of Pd nanoparticles, validating our nanocomposites for catalytic applications. The palladium-based catalyst was active for carbonylative coupling during 5 consecutive runs without loss of activity.

[1] N. Karousis, N. Tagmatarchis, D. Tasis, *Chem. Rev.* **2010**, *110*, 5366–5397.

[2] A. Desmecht, S. Hermans, O. Riant, *ChemistryOpen* **2017**, *6*, 231–235.

[3] A. Desmecht, D. Sheet, C. Poleunis, S. Hermans, O. Riant, *Chem. - A Eur. J.* **2019**, *25*, 1436–1440.

[4] A. Desmecht, T. Steenhaut, F. Pennetreau, S. Hermans, O. Riant, *Chem. - A Eur. J.* **2018**, *24*, 12992–13001.