Unraveling the Enhancement of the Interfacial Compatibility Between Metal-Organic Framework and Functionalized Graphene Oxide

Anusha Lalitha[‡], Jae Eun Shin[ζ], Satyanarayana Bonakala[‡], Jee Yeon Oh[ζ], Ho Bum Park[ζ,*] and Guillaume Maurin[‡,*]

[‡] Institut Charles Gerhardt Montpellier UMR 5253 CNRS, Université de Montpellier, Place E. Bataillon, 34095 Montpellier Cedex 05, France

[ζ] Department of Energy Engineering, Hanyang University, Seoul 04763, Republic of Korea.

Advanced modelling and experimental characterization tools were coupled to gain an unprecedented in-depth characterization of the interface formed when a hybrid porous solid ZIF-8 is incorporated as filler onto a pristine and an amine-functionalized graphene oxide (GO) matrix. As a preliminary stage, a pristine and an amino-pyridine functionalized GO was synthesized, and a realistic atomistic model was computationally built integrating their structural and chemical features deduced from X-ray photoelectron spectroscopy and X-ray diffraction experiments. An atomistic representation of the ZIF-8/functionalized GO interface was further simulated by a an innovative computational approach we developed and carefully analyzed in terms of the nature and the strength of interactions between the two components and the GO conformation with a special emphasis on the impact of the functionalization. It was revealed that grafting amino-pyridine function at the GO significantly enhances the interactions with the terminal functions present at the MOF interface associated with an *in*-depth penetration of the functionalized GO into the pockets of the ZIF-8. The so-predicted high compatibility between the two components was then supported by Infra-red experiments collected on the prepared composite. Complementary Transmission Electron Microscopy experiments further revealed a homogeneous dispersion of the ZIF-8 nanoparticles into the GO matrix with the absence of MOF agglomeration and mechanical testing evidenced a significant enhancement of the tensile strength for the corresponding composite. This fundamental exploration unambiguously demonstrates the key role played by the GO functionalization to achieve optimal interfacial MOF/GO properties and this opens promising perspectives for processing thin films required for future applications.