

Quantum Modelling and Electronic Structure Analysis in the Field of Materials Science

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Modelling based on various quantum chemistry codes is now part of the academic and industrial research landscape. The attraction of these simulation methods lies both in terms of the prediction of quantities that are sometimes difficult to extract at the experimental level for various materials whose interest is already well known, and in the possibility of exploring "unknown lands", upstream of synthesis work. In the more specific field of materials for energy or optics, it is now very well established that many key properties can be accessible by relying on this calculation tool, including in particular, structural and energetic characteristics related to the existence of point defects of more or less low content in the matrix, insertion/disinsertion phenomena, estimation of intercalation potentials in crystal systems or molecular redox potentials, activation energy of atomic jumps, etc. Even though a lot of information can be gathered directly by quantum chemistry calculation, already giving access to many possibilities of interpretation and also offering the possibility of opening the way to the transfer of this data in order to generate multi-scale models (*e.g.* in the field of matter transport phenomena that occur in batteries/fuel cells, etc.), the use of a more in-depth examination of the electronic structure can be very instructive. In many situations, this further step is of crucial help when searching for structure-property relationships.

This combination of approaches can be adopted in order to shed new light on Materials Science. Indeed, the route towards innovation and breakthrough can surely be accelerated thanks to the combination of quantum chemical modelling and methodologies belonging in particular to topological analysis of the electron density. In this area, the Quantum Theory of Atoms in Molecules ("QTAIM") method is of particular interest. Concerning materials for energy, while doping effects, oxygen/lithium or other metal (de-)insertion, effects of polymorphism, polyanion modulation, etc in inorganic matrices for M-ion batteries or Solid Oxide Fuel Cells traditionally constitutes one of the most prevalent playgrounds for computational studies, the more nascent field – in terms of development and applications – of batteries relying on organic electrode materials brings new perspectives and shifts the search in new directions. In this last context, the quest for most performant compounds is linked *e.g.* to new backbones identification, functionalization, isomerism, heteroatom substitution, redox centre change, etc. may benefits from the possibility to employ molecular modelling, prior to experiment, as a first estimation of their capabilities. By focusing on selective examples, the interest of quantum chemical modelling and topological analysis of the electronic structure applied to various compounds belonging either to organic or inorganic species will be illustrated in the purpose of driving the engineering search towards rationalization of phenomena and an educated guess of novel, innovating or optimized materials.

Keywords: Quantum Chemistry Calculations, Electronic Structure Analysis, Materials Characterization/Design, Structure-Property Relationships