The goal of the SOLAR2VAL project is to rationally design stable and highly efficient hybrid nanomaterials for heterogeneous catalytic applications, and namely for light harvesting/conversion and catalysis, to be implemented in proof-of-concept devices which can produce ammonia, ethylene or ethanol, by assembling a multidisciplinary and international team of computational chemists and physicists. The research project focuses on the study of the hybridization of selected nano-objects (the Nano Building Blocks, NBBs) and their assembly into a compact device. Key to this development is the control of the interactions and self-assembly properties of the NBBs at the nanoscale, in order to optimize the structure-property-function relationships in the integrated nano-systems, with the aim of mimicking the perfection of natural structures for light conversion and reach good efficiency and long-term stability in real devices.

The NBBs under investigation are indeed low-dimensional materials based on carbon, such as carbon quantum dots, two dimensional materials and a novel catalyst based on a bidimensional metal-organic framework structure, which are responsible for absorption of light, its conversion into energy and transfer towards the catalytic centre where the reaction can occur. The study will focus on the rational design of the assembling process among small, functional NBBs through a bottom-up approach, leading to integrated nanoarchitectures for highly efficient light harvesting/conversion and catalysis. Thus, the different NBBs will be coupled to each other to promote charge transfer (CT) processes which result into effective collection of charges at the catalytic center, to be able to perform the reaction to convert nitrogen into ammonia and CO_2 into ethylene and ethanol. To this end, the project relies on the rational design of the NBB components of the nanohybrids by means of multiscale computational modelling. Major efforts will be devoted to identify material design paradigms that can ensure limited recombination of charges/excitons at the nano-interfaces and at the same time enhance the CT processes for current generation, with the final goal of enhancing the efficiency of the catalytic process. The way the individual NBBs interact is thus crucial to ensure the realization of working devices, requiring a deep knowledge of the chemical and physical nature of the interface. Different levels of structure tuning will be considered in the hybrid, to enhance photocurrent generation and linking the morphology of the interface to its function: density ratio between components, relative distance, orientation and position, are all crucial aspects that will be considered.

In SOLAR2VAL, we will resort to multiscale computational approaches for the description of chemical and physical properties of the interfaces. This will be assessed considering different computational approaches in order to properly describe the light absorption process of the NBBs and the /CT at the interface. Moreover, it will be important for the description of the catalytic cycle to obtain the desired products. In particular, the time-evolution of the interface will be modelled, with a classical molecular dynamic method, which consist of considering the atoms bonded together in a spring-like way, without considering the electronic attractive forces explicitly. This allows to consider realistic model systems (thousands of atoms) and physically meaningful time-scales (hundreds of nanoseconds), for the investigation of the time-evolution of the interaction and stability of the different interfaces. Next, we will focus on accurate methods that account explicitly for the electrons, to describe and understand the electron and charge transfer mechanisms occurring at the interface and to optimize them, eliminating the charge recombination. Finally, the same methodology will be used to study the catalytic cycle, in order to understand the reaction mechanism and unveil the key reaction path which will lead to the formation of the desired products with high selectivity and efficiency.

SOLAR2VAL will develop a set of techniques for nano-structuring, with emphasis in delivering innovative rational design protocols for nano-hybrids from synthesis to processing, useful for the entire research community. The project will pursue a unique combination of cutting-edge materials, to create highly innovative platforms for optoelectronic and catalytic technologies. A considerable risk character describes its objectives, given the remarkable level of novelty that, in a single project, merges the modelling of different dimensionalities to obtain complex nano-hybrid architectures based on low-dimensional materials, as well as the testing of their functionalities. The high-reward of this research and innovation project is at hand, as it will sow the seeds of a new disruptive low-cost hybrid technology, stimulating the future implementation of new technological and economical assets in our future societies.