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## RESEARCH STATEMENT

Understanding that fossil fuels are not endless and that their extensive use is causing irreversible climate changes prompted us to realize that we are in urgent need of sustainable energy generation processes, energy vectors, and solutions to reduce pollution and greenhouse gas emissions. Despite replacing fossil fuels while maintaining or improving the current standards of living with a growing population is one of the biggest challenges that we have to face, the solution might lie in tiny pieces of matter: nanoparticles. Nanoparticles have been known for a long time but it is only recently that we have been able to better study and control their properties. The advent of nanotechnology and its associated tools allowed indeed to manipulate the composition, size, shape, functionalization and assembly of nanoparticles and to create nanoarchitectures and macroscopic devices with novel properties and unrivaled performance. In the Cargnello group, uniform and tailored nanoparticles and nanostructures are studied and used for energy and environmental applications, with emphasis on how to precisely control the nanoarchitectures to understand and exploit interactions between well defined building blocks. Applications include hydrogen generation through photocatalysis, reduction of methane emissions, pollution control, solar cells, to cite a few. It is expected that advancements in the preparation and use of these tiny particles can bring immense benefit for making big challenges more approachable.

Some of the research themes in the group are the following.

### ACTIVE AND STABLE CATALYSTS

Most of the industrial chemical transformations to prepare important building blocks are conducted at medium to high temperatures and pressures. Catalysts that are used to make these processes feasible have to be not only active, but also stable under reaction conditions for extended periods of time. Several mechanisms of deactivation are possible, the most severe involving the loss of active surface area either by particle agglomeration (sintering or ripening processes) or by poisoning by intermediates or byproducts. Reducing catalyst deactivation is therefore as essential as obtaining active catalysts.
The study of catalyst stability is central in the Cargnello group. Strategies for avoiding catalyst deactivation include the formation of core-shell structures, where the core active material is protected against sintering by a porous, thermally stable shell. Alternatively, the composition of the nanoparticles can be tuned so that poisoning and byproduct deposition is limited. Heterostructures, where two active compounds are placed close to each other, are also of interest for studying the physical interaction between two materials and their effect onto the final performance of catalysts.

*Related publications*

Cargnello, M.; Delgado Jaén, J. J.; Hernández Garrido, J. C.; Bakhmutsky, K.; Montini, T.; Calvino Gamez, J. J.; Gorte, R. J.; Fornasiero, P. "Exceptional activity for methane combustion over modular $Pd@CeO_2$ subunits on functionalized $Al_2O_3$.", *Science* **2012**, *337*, 713-717.

Cargnello, M.; Grzelczak, M.; Rodriguez-Gonzalez, B.; Syrgiannis, Z.; Bakhmutsky, K.; La Parola, V.; Liz-Marzan, L. M.; Gorte, R. J.; Prato, M.: Fornasiero, P. "Multiwalled Carbon Nanotubes Drive the Activity of Metal@oxide Core-Shell Catalysts in Modular Nanocomposites.", *J. Am. Chem. Soc.* **2012**, *134*, 11760-11766.

## PRECISE NANOCRYSTALS

The ability to tailor and tune matter at the very small scale is not only scientifically interesting, but also technologically very important. The discovery of new properties that make nanoparticles very different from their respective bulk materials is one of the most important achievements of nanotechnology. Forces and properties of nanoscale building blocks can indeed substantially deviate from the bulk because of the very small size of the structures. The energy levels in these tiny structures are intermediate between those of both molecules and bulk counterparts, with the possibility to tune their properties by tuning the size and shape of the building blocks. At the next level of control there is also the opportunity to combine uniform building blocks and obtain hierarchical architectures with an added degree of complexity. Self-assembly is therefore used as a tool to engineer complex structures for a specific task or to maximize interactions in multifunctional materials.

In this area, the Cargnello group explores the synthesis of new nanoscale building blocks and their assembly into hierarchical structures that allow the communication (catalytic or electronic) between different materials and phases. The interactions are controlled at the nanoscale level to allow flow of energy or matter from one component to another, leading to controlled and improved properties.

*Related publications*

Cargnello, M.; Diroll, B. T.; Gaulding, E. A.; Murray, C. B. "Enhanced energy transfer in quasi-quaternary nanocrystal superlattices." *Adv. Mater.* **2014**, *26*, 2419.

Cargnello, M.; Doan-Nguyen, V.; Gordon, T. R.; Paik, T.; Diaz, R. E.; Stach, E. A.; Gorte, R. J.; Fornasiero, P.; Murray, C. B. "Control of Metal Nanocrystal Size Reveals Metal-Support Interface Role for Ceria Catalysts.", *Science* **2013**, *341*, 771.

## PHOTOCATALYSIS

The Sun is an (almost) infinite and costless source of energy that provides us with an amount of energy far exceeding our needs. Plants and some organisms have learnt during millennia of evolution to exploit this unique privilege to live on our planet. The use of this clean and sustainable energy source would allow us to solve most of the problems connected to the utilization of fossil fuels. There are however issues associated with solar energy, mainly related to the fact that it is a diffuse and intermittent source of energy. Harvesting and storing solar energy are therefore some of the key problems to overcome to achieve a truly sustainable energy scenario. Nanostructures are natural candidates for this role, because of their high surface-to-volume ratio that increases their capability to harvest solar radiation and because of their tunable properties that permit manipulation of their energy levels. In analogy to how carotenes and chlorophylls harvest and transfer solar energy in plants, arrays of nanostructures can play this role and funnel energy to reactive centers for the transformation of solar into chemical energy in the form of chemical bonds. To this end, the production of liquid compounds where the energy is stored is one of the most attractive solutions.

Studying photocatalytic processes that turn solar energy into chemical energy is an important objective in the Cargnello group. Fundamentals of photocatalytic processes are studied by using uniform and tailored nanostructures based on abundant semiconductors. The controlled positioning of building blocks is key to collect and transfer energy to reactive centers that are then capable of transforming abundant molecules into liquid compounds that can be used as fuels. To this scope, important compounds such as hydrogen (as precursor for methanol synthesis) and methanol are the most interesting targets.

*Related publications*

Gordon, T. R.; Cargnello, M.; Paik, T.; Mangolini, F.; Weber, R. T.; Fornasiero, P.; Murray, C. B. "Nonaqueous Synthesis of $TiO_2$ nanocrystals Using $TiF_4$ to Engineer Morphology, Oxygen Vacancy Concentration, and Photocatalytic Activity.", *J. Am. Chem. Soc.* **2012**, *134*, 6751.