

The Research Group
General Chemistry

has the honor to invite you to the public defence of the PhD thesis of

José Luis Nuñez

to obtain the degree of Doctor of Sciences

Joint PhD with Universidad Nacional del Litoral

Title of the PhD thesis:

**An atomistic view point of the chemical reactivity of
modified carbon materials**

Supervisors:

Prof. dr. Frederik Tielens (VUB)

Prof. dr. Paola Quaino (UNL)

The defence will take place on

Tuesday, March 18, 2025 at 1 p.m. (CET)
in "Aula Leloir", FIQ, UNL, Argentina.

The defence can also be followed through a live stream:

Microsoft Teams

Meeting ID: **360 897 996 506**

Access code: **BS6qL2vP**

Members of the jury

Prof. dr. Frank De Proft (VUB, chair)

Prof. dr. Pablo Bolcatto (UNL, secretary)

Prof. dr. Mercedes Alonso Giner (VUB)

Prof. dr. Alfredo Juan (UNS)

Curriculum vitae

José Luis obtained his 'Licenciatura en Química' degree at the Universidad Nacional del Litoral (UNL), Argentina, in April 2020, after presenting a degree thesis on the ab initio studies of modified carbon nanotubes and OER reaction. He also worked on experimental chemistry at the Physics Department (FBCB, UNL, Argentina), particularly in EPR characterization of a copper dianionic complex.

He is interested in the use and development of computer codes applied to the description and prediction of chemical properties of novel materials, which better understanding can help science and technology advancement.

Abstract of the PhD research

One of the ways to achieve a reduction in the use of high-cost catalysts is through the synthesis of new compounds with low proportions of these catalysts, which maintain or enhance their desirable characteristics. Conversely, carbonaceous materials offer large surface areas with diverse geometries and compositions that can serve as supports at a significantly reduced cost. However, a better understanding of the synthesis conditions of these systems is still required in order to control their preparation and characteristics and, in particular, the changes in their catalytic properties that may arise from the synergistic effect of the combination of these materials.

This thesis aims to address theoretical studies that provide insight into the behavior of carbonaceous materials modified with nanoparticles of a metal or oxide in different structural configurations and compositions. The objective is to develop a comprehensive understanding of how changes in the material properties after modification impact water adsorption and dissociation. Based on these findings, the goal is to establish a solid foundation for the design of new materials with enhanced performance for the studied processes. For this matter, a series of chemical models of hybrid materials have been suggested and investigated through the use of ab-initio methods. In particular, two forms of carbon surfaces, namely carbon nanotubes (CNT) and graphene oxide (GO), have been proposed as substrates for the deposition of different platinum and iridium oxide nanoparticles.

As a result of this investigation, it is found that the proposed carbon surfaces can stabilize platinum (Pt_n , $n = 1-10, 13$) and stoichiometric iridium oxide ($(\text{IrO}_2)_n$, $n = 1-6$) nanoparticles. The small diameter of the CNT clearly limits the interaction area between the particles and the carbon surface. Moreover, iridium oxide "core-shell-like" structures are found to be more suitable for interacting with polar surfaces as that of the GO. Furthermore, water adsorption and further dissociation were analyzed on selected Pt_n/CNT and $(\text{IrO}_2)_n/\text{CNT}$ systems. Both types of systems presented outstanding results regarding activation energies for the dissociation step. The results of this study encourage experimentalists to synthesize stable triangular-shaped Pt nanoparticles and nanoscaled iridium oxide particles on a CNT surface to promote polarization and increase the efficiency of catalysis.

Lastly, this thesis further examines M_{13} monometallic nanoparticles and their reactivity (by using simple molecules, such as O_2 and N_2), with the aim of developing a comparable analysis for M_{13} clusters on carbon-based surfaces in the future.