Combined theoretical-experimental investigation of metal-based interfaces: from catalysts to nanofluids

Dr Ricardo Grau-Crespo

Department of Chemistry, University of Reading, Whiteknights, Reading RG6 6DX, UK.

Abstract: The combination of density functional theory (DFT) simulations and experimental characterization techniques provides a powerful tool for the investigation of metal-based interfaces. In this talk, I will discuss computer-modelling work done in my group, in collaboration with experimentalists, about the properties and interactions of metal surfaces. The Re/Pd interface, which is largely unstrained, provides a useful playground to test the agreement between X-ray photoemission spectroscopy (XPS) measurements and density functional theory (DFT) calculations of core levels, and to assess the role of interfacial charge transfer in the core and valence electronic structure [1]. Using a combination of DFT, XPS and NEXAFS (near-edge X-ray absorption fine structure), we have studied the interaction of nickel surfaces with chiral modifiers and other organic molecules, to gain insights on the use of modified nickel as an enantioselective catalyst [2-4]. I will illustrate how the combination of synchrotron-based techniques and simulations allows the unambiguous determination of adsorption geometries in a way that is not accessible to experimental or theoretical techniques working in isolation. Finally, I will highlight our recent work, again in collaboration with experimentalists, examining the role of metal-organic interfaces on the thermal properties of nanofluids that are of interest as heat transfer fluids in concentrated solar power plants [5-8].

1. Ontaneda, J., R.A. Bennett, and R. Grau-Crespo, *Electronic Structure of Pd Multi layers on Re(0001): The Role of Charge Transfer.* Journal of Physical Chemistry C, 2015. **119**(41): p. 23436-23444.
2. Ontaneda, J., et al., *Adsorption of Methyl Acetoacetate at Ni{111}: Experiment and Theory.* Journal of Physical Chemistry C, 2016. **120**(48): p. 27490-27499.
3. Tsaousis, P., et al., *Combined Experimental and Theoretical Study of Methyl Acetoacetate Adsorption on Ni{100}.* Journal of Physical Chemistry C, 2018. **122**(11): p. 6186-6194.
4. Quevedo, W., et al., *Adsorption of Aspartic Acid on Ni{100}: A Combined Experimental and Theoretical Study.* Langmuir, 2020. **36**(32): p. 9399-9411.
5. Carrillo-Berdugo, I., et al., *Understanding the Specific Heat Enhancement in Metal-Containing Nanofluids for Thermal Energy Storage: Experimental and Ab Initio Evidence for a Strong Interfacial Layering Effect.* Acs Applied Energy Materials, 2020. **3**(9): p. 9246-9256.
6. Carrillo-Berdugo, I., et al., *Optical and Transport Properties of Metal-Oil Nanofluids for Thermal Solar Industry: Experimental Characterization, Performance Assessment, and Molecular Dynamics Insights.* Acs Sustainable Chemistry & Engineering, 2021. **9**(11): p. 4194-4205.
7. Carrillo-Berdugo, I., et al., *Interfacial molecular layering enhances specific heat of nanofluids: Evidence from molecular dynamics.* Journal of Molecular Liquids, 2021. **325**.
8. Carrillo-Berdugo, I., et al., *Interface chemistry effects in nanofluids: Experimental and computational study of oil-based nanofluids with gold nanoplates.* Journal of Molecular Liquids, 2022. **362**.