ABSTRACTS - CONFERENCES

C1. PROCESS INTENSIFICATION FOR THE CLEAN OXIDATION OF ALCOHOLS

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The postulates of green chemistry promote “clean” technologies that use renewable feedstock together with safer solvents and auxiliaries to develop less hazardous chemical syntheses. Clean technologies should have robust process conditions to sustain long periods of operation with minimal intervention, low waste generation, simple downstream purification and the production of green and sustainable products. Oxidation of alcohols is vastly applied in chemical synthesis to get other chemicals and end-products, like solvents, fine chemicals, food additives, pharmaceuticals. Oxidation routes developed in continuous flow, using air or oxygen as oxidants and water without additives as the solvent, and preferably at moderate operating conditions are candidates of succeeding as clean technologies. Process intensification (PI) is the strategy of making improvements in processing that substantially decreases equipment size, waste production and/or energy consumption, resulting in more efficient chemical plants. In the presentation, the concept of PI will be introduced, and some results related to the clean oxidation of alcohols will be shown.
C2. HOW A SMALL CHANGE CAN MAKE A BIG DIFFERENCE: MODIFYING THE LIGAND SPHERE IN SULFUR-CHELATED RUTHENIUM BENZYLIDENES

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For the past decade we and others have disclosed the potential of developing sulfur-chelated ruthenium benzylidenes to advance latent olefin metathesis and chromatically orthogonal sequences.[1] Recently, we have shown that changing the NHC framework, and even replacing the NHC by CAAC ligands has a significant influence on the activity and the selectivity of these catalysts. Some of these changes brought about the expected results;[2] while others were quite surprising.[3] However, exchange of the anionic ligands[4] in these complexes certainly resulted in the most unexpected selectivity with great practical implications in the field of olefin metathesis.[5]

References.
C3. HNO COMPLEXES OF METALLOPORPHYRINS

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A Co-porphyrin modified electrode was developed, and the three-electrode sensor arrange detects down to 1 nM [HNO]. The sensor discriminates HNO from NO, and responds to repeated additions of an HNO donor, without signal loss. Moreover, the detection can be achieved in real time (is time-resolved) i.e., the sensor has a fast response to changes in [HNO], so that the instant concentration of HNO can be monitored. Recent evidence suggests that HNO could be involved in biological processes, some of which are attributed to NO. In this context, one of the most important and yet unanswered questions is whether HNO is produced in vivo. Possible routes concern chemical or enzymatic reduction of NO. Experiments with the sensor show that HNO is produced from NO by alcohols with moderate reducing capacity, such as the biologically relevant vitamin C .Other common aromatic alcohols obtained from foods, such as Vitamin E, or used as over-the-counter drugs, like aspirin, are able to undergo the reaction. The proposed mechanism involves nucleophilic attack to NO by the alcohol, coupled to proton transfer and subsequent decomposition of the so-produced radical to yield HNO and an alkoxyl radical. Aliphatic and aromatic thiols (as well as selenols) are also able to convert NO to HNO, albeit at different rates. Further mechanistic analysis using ab initio methods shows that the reaction between NO and the thiol produces a free radical adduct RSNOH•, which reacts with a second NO molecule to produce HNO and a nitrosothiol. The nitrosothiol intermediate reacts further with RSH to produce a second molecule of HNO and RSSR, as previously reported. Quantum mechanical calculations and Monte Carlo statistical mechanical simulations were carried out to evaluate the proton-coupled one electron reduction potential of NO to HNO, \( E'' = -0.161 \) V.\(^{(6)} \) The results show that the process can be promoted by well-known biological reductants such as NADH, ascorbate, vitamin E, cysteine, and glutathione, for which the reduction potential at physiological pH is around \(-0.3\) to \(-0.5\) V. The computed reduction potential of NO through the radical anion HONO•−, which is involved in equilibrium with NO in aqueous solution, can also explain the recent experimental findings on the formation of HNO through the reduction of NO, promoted by H2S, vitamin C, and aromatic alcohols. Finally, Fe porphyrins have been used to stabilize coordinated HNO by chemical or electrochemical reduction of \( \{MNO\}_7 \) porphyrins to \( \{MNO\}_8 \). Studies which were done previously in organic solvents, and using metalloporphyrins with electron-attracating groups in order to stabilize NO– coordinated to Fe, are nowadays being reproduced in aqueous solutions without electron-attracting groups. Probably, H2O stabilizes the HNO/ NO– ligand through H-bonding.

References.
C.4. EMERGENCE OF FUNCTION IN PRIMITIVE CHEMICAL NETWORKS OUT OF EQUILIBRIUM

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Like many other open systems in nature, living organisms are replete with rhythmic and oscillatory behaviour at all levels, to the extent that oscillations have been termed as a defining attribute of life. Recently, we have started to investigate a key challenge in contemporary Systems Chemistry research,[1] that is, to synthetically construct “bottom-up” peptide-based networks that display bistable behaviour and oscillations. Towards this aim, we utilize replicating coiled coil peptides, which have already served to study emergent phenomena in complex mixtures. In the first part of this talk, we describe the kinetic behaviour of small networks of coupled oscillators, producing various functions such as logic gates, integrators, counters, triggers and detectors.[2] These networks are also utilized to simulate the connectivity and network topology observed for the Kai-proteins circadian clocks, producing rhythms whose constant frequency is independent of the input intake rate and robust towards concentration fluctuations.[3] Then, in the second part, we disclose our experimental results, showing for that the peptide replication process can also lead to bistability in product equilibrium distribution.[4-6] We believe that these recent studies may help further reveal the underlying principles of complex enzymatic processes in cells and may provide clues into the emergence of biological clocks.

References.
C5. QUANTUM TUNNELING INSTABILITY - WHEN SYNTHESIS IS FUTILE
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One of the hardest, but also most rewarding task in chemistry consists in the generation of any molecule we can envision. As Hoffmann stated “…the literature of organic chemistry has contained characterizations of molecules as unstable, strained, distorted, sterically hindered, bent and battered. Such molecules are hardly seen as dull; on the contrary, they are perceived as worthwhile synthetic goals, and their synthesis, or evidence of their fleeting existence, acclaimed. What is going on here? Why this obsession with abnormal molecules? Is this molecular science sadistic at its core?” This raises the question: is it even in the realm of possibilities to generate all those extreme molecules? On this matter computational chemistry has the upper hand: it can predict with high confidence the stability of any system whether in kinetic or thermodynamic terms. Moreover, it can predict the probability of decomposition of such molecules by a quantum mechanical tunnelling mechanism in what we can call “Quantum Tunnelling Instability” (QTI). In this talk we will discuss the preliminary analysis of several cases of “impossible molecules”, systems which should be synthesizable and stable at low temperature, but their half-lives are in fact extremely short due to unavoidable quantum tunnelling effects (from the ground state or in thermally activated tunnelling).
C6. SYNTHEtic OLigosacharides OF Pathogenic Microorganisms FOR THE DEVELOPMENT OF Glycobiology

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Complex carbohydrates are involved in a wide range of biological processes, which encompass infection, metastasis, cell signaling, cellular proliferation, cellular adhesion, pathogen recognition, inflammation, etc. Consequently, glycobiology has turned into an area of intense research. A major obstacle for its development is the lack of well-defined oligosaccharides, which are difficult to isolate in pure form from natural sources. The chemical synthesis of oligosaccharides often emerges as a requirement and involves the construction of glycosidic linkages regio- and stereoselectively. A donor and an acceptor have to be carefully chosen considering the influence they exert on the stereochemical course of the glycosylation reaction. Among the oligosaccharides found in pathogenic microorganisms, glycans containing galactose in the furanose form (Galf) are quite relevant because mammals only have galactose in the pyranose form (Galp). Several D-Galf-glycoconjugates from pathogenic microorganism proved to be essential for viability or survival which prompted to consider the Galf-metabolism as a potential target of chemotherapy.
C7. CHEMICAL INFORMATION EXCHANGE AS A REGULATOR OF WAR AND LOVE BETWEEN SPECIES

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Life on earth is heavily based on chemical communication between cells. Quorum sensing enables unicellular organisms to coordinate their behavior and function in such a way that they can adapt to changing environments and compete, as well as coexist, with multicellular organisms. Prime examples of this phenomenon are displayed by the opportunistic pathogens Pseudomonas aeruginosa and Agrobacterium tumefaciens, which cause disease in immunocompromised humans and plants, respectively. Quorum sensing in these pathogens is mediated by small amphiphilic signaling molecules such as 3-oxo-C12-HSL and 3-oxo-C8-HSL, leading to biofilm formation and secretion of virulence factors. The Meijler group is targeting QS in various pathogens with several chemical tools, such as a set of electrophilic probes that are designed to bind QS receptors covalently, leading to inhibition of QS regulated gene expression. These probes are used as molecular tools to obtain new insights into the mechanisms of activation and deactivation of bacterial quorum sensing, as well as plant hormone signaling. Furthermore, it was recently found that certain QS molecules and other natural products can also directly affect the behavior of other bacterial species as well as that of eukaryotes. Diverse eukaryotes have been found to react strongly to the presence of these compounds (often initiating counter-warfare to jam bacterial communication), however, to date no eukaryotic receptor has been identified that specifically binds bacterial QS molecules. The Meijler group has synthesized and used a set of `tag-free` probes to isolate and identify for the first time such receptors.
C8. NARROW-SPECTRUM ANTIBACTERIAL AGENTS, DEVELOPMENT OF NEW INHIBITORS OF MmpL3 FLIPPASE IN MYCOBACTERIA

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The search for new chemotherapeutics against bacterial infections is one of the most successful stories in human therapeutics of the 20th century. This has culminated in the discovery of several families of antibiotics that have allowed to drop the death rates from infectious diseases. Nature has played a central role in this context with the isolation of several natural products, which turned to be useful leads for the development of drugs. However, the emergence of new generations of antibiotic-resistant pathogen has eroded the effectiveness of current drugs and constitutes today a major threat to public health, infectious diseases being the 3rd leading cause of death in developed countries. Our research group uses as inspiration the structures of bioactive natural organic compounds as a starting point for the search for new chemical entities with therapeutic action. Using the structure of the cinnamic acids as a template, synthetic modifications and studies of the structure activity relationship as antimicrobials were carried out. Different families of compounds were tested against fourteen different bacterial strains finding selective inhibition of the mycobacterial genus.

References.
C9. CHEMICAL BIOLOGY AND PRECISION MEDICINE

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Next Generation Sequencing technologies have driven biology in the big-data paradigm, with genomic information growing exponentially on a daily basis. Bioinformatics is the core business that allows conversion of this information into biological knowledge, and among most important issues are genotype-phenotype inference relationships. Predicting how genetic differences impact a given protein function at the molecular level, and thus phenotypic outcome, is important for understanding among other issues: antibiotic resistance and development of new drugs, enzyme selectivity and specificity, and human genetic borne diseases. In the present talk I will show examples of several in-house developed bioinformatic applications that allow structure based physico-chemical characterization of proteins that allow prediction of whether a newly found human mutation is likely to be pathogenic.
C10. WHAT MAKES ELECTROACTIVE BACTERIA, ELECTROACTIVE?

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One of the research questions that we have found most fascinating in bioelectrochemistry is, what makes electrogenic bacteria, electrogenic? And more importantly, can we turn non-electrogenic bacteria to electrogenic bacteria? Using the tools of synthetic biology, synthetic chemistry and standard as well as new genetic engineering tools, we have replaced the pili of pseudomonas aeruginosa with pili of electrogenic bacteria using CRISPR/Cas 9 technology and studied the new bacteria properties. We have genetically expanded the genetic code of pseudomonas aeruginosa and modified its flagellin peptides using an unnatural amino acid (UAA) in order to site-specifically attach to it electro-transferring enabling moieties (such as gold nanoparticles). Using these novel technologies, we could successfully confirm some of the suggested mechanisms of electron transfer in such electron transferring microorganisms, furthermore we could successfully demonstrate that if specific molecular components are correctly identified as electron transferring components, transplantation of these proteins into non-electron transferring microorganisms, greatly enhances their electron transferring properties.
Breast Cancer (BC) is a complex disorder due to multiple genes deregulation, which prompts a robust phenotype. The pharmacological paradigm of “magic bullets”, targeting individual chemoreceptors, fails as redundant functions are activated and alternative compensatory signaling routes sustain the tumor phenotype, leading to immune escape, chemoresistance and metastasis. In this way, polypharmacology arises as a new paradigm: designing multifunctional drugs that interact with several molecular targets. This approach would tackle several signaling and metabolic routes at the same time, with one drug, leading to new and more effective treatment against BC. Our hypothesis is that a dual antagonist of both, Estrogen Receptor (ER) and Liver X Receptor (LXR), would inhibit the ER canonical survival routes, but also would inhibit lipogenesis and Warburg effect through LXR antagonism. These two are key metabolic pathways that drive cancer progression, growth, survival, immune evasion, resistance to treatment and disease recurrence.

In this sense, we performed a screening of different natural and synthetic oxysterols by reporter gene assay on both, ER and LXR. We studied their binding mode and molecular dynamics simulations to gain insight on the molecular determinants involved in their activity. Compound 1 emerged as a dual ERα/ERβ antagonist at micro molar concentration, and a dual LXRα/LXRβ inverse agonist at micro molar concentration, in reporter gene assays. 1 effectively inhibits proliferation and migration on the ER+ BC cell line MCF7 and the ER- BC cell line MDA-mb-231. Moreover, it inhibits migration of the human vascular endothelial cell line EA.hy926, by suppressing the NFκB signaling pathway, suggesting anti-angiogenic activity.
C12. FUNCTIONAL FOOD NANOCOLLOIDS

Ana M. R. Pilosof

Food nanocolloids are nano-sized materials within a solvent that is always water, that use safe edible materials. Examples of engineered food nanocolloids include nanoemulsions, nanomicelles, nanocapsules, nanofoams, nanoliposomes, nanogels, nanofibers. We will focus on functional nanoemulsions that have considerable potential as carriers of bioactive components with specific health benefits. They may be designed specifically for protecting and delivering via the oral route lipophilic bioactive components. However, the contact with conditions of gastric fluids, with low pH, high concentrations of salts and ionic strength, and the often low solubility during gastrointestinal digestion may result in only a small proportion of bioactive molecules remaining available which limits the activity and potential health benefits of bioactive compounds. As caseinomacropeptide (CMP), a random coil peptide that comprises the 64 amino acids in the hydrophilic C-terminal portion of kappa-casein has the ability to self-assemble into different types of nanostructures in solution and at oil/water interfaces, it has a potential role to produce smart nanosized emulsions. We will discuss how they can be stabilized to undergo a pH-dependent gelation, that assures a high bioaccessibility upon digestion.
This talk will focus on two classes of polymers that were designed for sustainability. The first class is called single chain polymeric nanoparticles (SCNPs). These are water-soluble polymers that fold into a compact structure that loosely resembles a protein. However, its structure is less well defined but it can be engineered to exhibit similar and significantly broader activities. Indeed, we have integrated in one or more transition metal catalysts within the SCNP structure so that the overall nanoparticle performs like a metalloenzyme. A portion of the talk will focus on characterizing these nanoparticles, demonstrating their enzyme-like properties, and exploring structure activity relationships with a range of substrates. Beyond performing catalysis in water, an environmentally friendly solvent, biological applications can be envisioned for these nanoparticles and at least one example will be given. The second part of the talk focuses on the development of degradable polyurethanes for end-of-life repurposing. Millions of tons of polyurethanes (PU) are produced every day. The widespread commercial success of PUs is in large part because of their low cost, facile synthesis, and attractive physical properties. Thus, a wide range of polyols and polyisocyanates undergo poly-addition polymerization to produce elastomers, foams, adhesives, and coatings for a myriad of applications. Due to PU’s mass production globally, it is no surprise that there is an enormous amount of PU waste generated daily. PU waste has become a huge global concern since most are disposed into landfills or the ocean, harming aquatic wildlife. This talk will discuss on new class of PUs that can be degraded down to slightly modified polyols and then repurposed into new materials. Importantly, the degradation can be performed at room temperature with an organic solvent and an organic acid.
C14. DEVELOPMENT AND CHARACTERIZATION OF NANOSTRUCTURED SURFACES PRESENTING AFFINITY TO HEAVY METAL IONS.

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The development of sensitive and specific chemical systems on metallic platforms is of great interest due to their potential applications in sensor, biosensor, electrocatalysis and molecular electronics among others. A precise control of the metal-organic interface is needed to design useful devices. Self-assembled organic monolayers (SAMs) such as thiols have been shown to be the most promising systems to modify the surfaces of well-defined, rough, nanoparticle, nanorod, nanoshell or nanocage gold substrates because they provide an organic platform of controllable molecular thickness able to anchor different species by nonspecific interactions.

In the preparation of ionic sensors, organic molecular ligands are widely used due to its specific recognition for metals. There is a correlation between behavior and macroscopic, chemical and physical properties of modified electrodes. For a clever design of the electrode modification, it is of great importance the physicochemical characterization of the systems and their behavior so that this will lead to an optimization of the surfaces according to the process of modifying them and their expected use.

We will present results of studies of functionalized gold surfaces with different organic functional groups with oxygen as donor (-COOH, -OH). We have tried their affinity with copper ions and have characterized the surface-ion interaction via electrochemistry, X-Ray Photoelectron Spectroscopy and Infrared Spectroscopy. Experiments show that copper binds both to alcohol and carboxylic acid terminated SAMs, and in similar amounts. In both cases the cation interacts with the terminal functional group, and does not intercalate into the alkyl chain.

In the case of carboxylic acid functional group, this interaction causes the deprotonation of the acid and consequent formation of the salt. Different ways of coordination of the copper with the carboxylic acid moieties are possible so that in the electrochemistry analysis, three signals at different reduction potentials with not reproducible intensity are observed. In the case of alcohol functional group, the reduction potential is well defined.

With this information in mind, a polyacrylic acid (PAAcid) immobilized in a Nafion® polymeric matrix on graphite screen printed electrodes for detecting copper was prepared and tested, showing similar results.
C15. NANOMATERIALS AT INTERFACES: WET CHEMICAL ROUTES FOR SIZE, SHAPE AND PHASE - CONTROLLED NANOMATERIALS

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Wet chemical routes for synthesis of semiconducting materials are straightforward, cost-effective and often result in high quality nanomaterials with precise size and shape control. Two synthetic methods will be presented, both in which interfacial processes play a major role. The first method, chemical solution deposition, offers a simple and versatile route for producing high quality semiconductor thin films directly onto single crystal substrates without the use of organic ligand molecules. A wide range of microstructures is obtained, from nanocrystalline films to ‘chemical epitaxy’ - monocrystalline thin films with a well-defined orientation with respect to the substrate. The second part will present the synthesis of highly uniform nanoparticles capped with alkylamine surfactants, focusing on the role of ‘beneficial impurities’ for controlling their shape and phase.
C16. MOLECULAR NANOMAGNETS BASED ON 3D/4F COORDINATION COMPLEXES

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Over approximately the last fifteen years, the field of Molecular Magnetism shifts the focus away from polynuclear complexes toward single-ion based ones, particularly pointing to those containing lanthanide ions [1]. It comes out that for f-element complexes (lanthanide based), spin–orbit coupling is much stronger than crystal field effects (especially for 4f elements), and as consequence, the total angular momentum, J, is used to classify the states of the low energy manifold. In this scenario, the thermal barrier to relaxation, $U_{\text{eff}}$, is determined by the energy splitting of the $m_J$ microstates of the ground J multiplet. The systematic study of a family of Ln(III) complexes as simple as possible, appears as an interesting approach to further understand slow relaxation of magnetization including QTM mechanisms and even other mechanism that are well known to be operative in these systems as Raman or Direct relaxation ones[2]. If the influence that magnetic interaction exerts in Ln(III) based SMMs is likely to be analyzed, then dinuclear Ln(III)-Ln(III) systems must be chosen, in fact several reports can be found about this type of complexes [3]. The dinuclear systems based on the Co2IIILn2III unit has been developing during last year’s [4]. As low spin Co(III) is a diamagnetic ion, this type of compounds, from magnetic point of view, are genuine dinuclear lanthanide ones. Moreover, the inclusion of the Co(III) coordination spheres, offers more synthetic versatility at the moment of fine tuning the crystal field around the Ln(III) sites.

This talk will focus on some selected Co2IIIDy2III systems discussing synthetic strategies, structural diversity and magnetization slow relaxation behavior, mainly emphasizing structural and electronic factors and their relationship with SMM properties.

References
C17. PEPTIDE BASED BIOELECTRONICS

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Billion of years of natural evolution has resulted in an enormous amount of proteins that are involved virtually in any biological process. Mimicking the unique features of these effective natural machineries, can be extremely productive for advancing nowadays technologies. Motivated by this approach, our group utilizes de-novo designed peptides that capture many of the proteins features but are also optimized for electronic applications. This approach will be demonstrated in this talk using three examples applicable for: electron and proton transport,1-4 small molecules sensing at the single molecule level,5 and modulating surface electronic properties of inorganic semiconductors.6-7 These examples demonstrate that de-novo designed peptides can be powerful building blocks for the preparation of novel, high performance, and biocompatible organic and hybrid bioelectronic materials, fabricated by cost effective and environmentally friendly methods.

References
The analysis of chlorophyll a fluorescence in plants allows obtaining relevant information about the photosynthetic activity. The key that connects photosynthesis with fluorescence is provided by the competitiveness of the processes that lead to the deactivation of chlorophyll, once it is excited by light absorption or energy transfer. In fact, there are three competing processes: i) transfer of electrons from the excited state, which initiates the electronic transport of the photosynthetic chain, ii) heat dissipation and iii) emission of fluorescence. This talk presents a review of the methodologies used in the analysis of chlorophyll a fluorescence for diverse photosynthetic materials; both at laboratory and field level, and physical models are explained to correct distortions by light re-absorption processes. It introduces how information on the effect of pollution and/or environmental stress on the health of crops can be obtained from the analysis of the fluorescent emission. The main achievements of our research group are summarized, both for natural media and for hybrid systems constituted by photosynthetic material with incorporated nanoparticles.
C19. STRATEGIES FOR SUSTAINABLE CONVERSION OF BIOMASS INTO BIOFUELS AND VALUABLE PRODUCTS FOR ENVIRONMENTAL MITIGATION

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Among recent strategies explored in our research group to improve yield and quality of the liquid products (bio-oils) arising from pyrolysis of lignocellulosic biomass for potential use as biofuels, results attained by minerals removal from selected biomasses through mild acid pretreatment are presented. Pyrolysis assays performed in a bench-scale installation for the raw and demineralized biomasses enable to obtain and characterize the three kinds of pyrolysis products, namely bio-oils, an enriched carbon solid product (bio-char), and gases, as well as to examine their potential applicability tending to promote process sustainability and biomass-based pyrolysis poly-generation technology. On the other hand, conversion of biomass into activated carbons by applying microwave assistance and hydrothermal carbonization, instead of conventional heating and carbonization, respectively, is described, including the adsorptive behavior of the resulting carbons in the removal of representative water contaminants. Recent advances targeted at the development of highly efficient activated carbons for CO2 adsorption mimicking post-combustion conditions, as an alternative to overcome drawbacks related to the absorption technology for post-combustion applications, are also summarized. Finally, mention is made of our achievements in the synthesis and potential applications of carbon nanotubes and graphene oxide nanosheets. They focus on possible use of the carbon nanotubes for H2 sensing at room temperature and transdermal delivery of therapeutic molecules, passive and electromodulated, as well as on post-combustion CO2 capture by graphene oxide sponge, and alginate beads loaded with graphene oxide nanosheets as nanostructured adsorbents of water contaminants.
Li-ion batteries have become the leading technology in energy storage to enable portable electronics and electric vehicles, hence the sustainable extraction of lithium from natural sources is of great technological as well as academic importance. We have developed an electrochemical method to extract LiCl from the natural brines present in the northwest of Argentina. The method uses an electrochemical reactor composed of Li+ and Cl- reversible electrodes. In this presentation I will discuss the method highlighting its working principle and main challenges to overcome.