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O₂ reduction at MN₄ macrocyclic complexes pentacoordinated through pyridine grafted on carbon nanotubes

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Metallophthalocyanines and metalloporphyrins are well known electrocatalysts for the reduction of O₂ (ORR) and have been extensively investigated with the aim of a better understanding of the ORR at modified electrodes and with the final scope of replacing Pt in fuel cells. Inorganic catalysts mimicking the structure of active redox centers of redox enzymes are being developed. For example Cho et al. reported higher electrocatalytic activity than Pt/C catalyst for a Fe-phthalocyanine with an axial ligand anchored on single walled carbon nanotubes that mimics the active site of cytochrome c oxidase. Fundamental studies of MN macrocyclic complexes adsorbed on carbon nanotubes (CNTs) and on pyridine modified CNTs as catalysts for the ORR are missing. Preliminary studies in basic media show that when MN macrocyclic complexes are adsorbed at CNTs (single walled and double walled) modified electrodes an increment of almost two orders of magnitude of catalytic activity expressed as log i at constant potential is observed. The reaction mechanism deduced from Tafel slopes analysis appears to be the same among the MN₄ complexes adsorbed and non-adsorbed on CNT, suggesting that the CNTs support does not interfere with the reaction mechanism. An increased electroactive area and therefore an increased amount of active sites and an increment of the formal potential of the catalyst seem to be the cause of this increment. The results show the same half-volcano trend reported before where the activity increases as the formal potential of the metal catalyst redox center is shifted to more positive values. When CNTs are modified with 4-aminopyridine aryl diazonium salts and then with axially ligated phthalocyanines even higher activities are observed but volcano correlations are distorted by the effect of the axial ligand.

Biography

Federico Tasca has completed his PhD studies in 2010 at Lund University under the supervision of Professor Lo Gorton and is working with bioelectronics and carbon nanomaterials. He continued his Post-doctoral studies in the Department of Chemistry at Stanford University. He is currently working as Assistant Professor at the Department of Chemistry of Materials of the University of Santiago of Chile. He is the Director of one Fondecyt Grant and of one Dycit Grant. He has published more than 25 papers in reputed journals in the field of Electrochemistry, Electrocatalysis Coordination Chemistry and Bioelectrocatalysis.

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