Recent advances in first principles based modeling and simulations of the physics and chemistry of large, complex atomistic systems

WILLIAM A. GODDARD, III

Charles and Mary Ferkel Professor of Chemistry, Materials Science, and Applied Physics Director, Materials and Process Simulation Center (MSC) California Institute of Technology (139-74) Pasadena, CA 91125

Advances in theoretical chemistry, computational chemistry, materials science, physics, and supercomputers are making it practical to consider first principles (de novo) predictions and simulations of the atomistic level physics and chemistry of complex systems and processes in the Chemical, Biological, and Materials Sciences. Our approach is to build a hierarchy of models each based on the results of more fundamental methods but coarsened to make practical the consideration of much larger length and time scales. Connecting this multi-paradigm multi-scale hierarchy back to quantum mechanics enables the application of first principles to the coarse levels essential for practical simulations of complex systems.

We will highlight some recent advances in multi-paradigm multi-scale methodology selected from: · The ReaxFF reactive force field for prediction of reactive processes in complex systems · The eFF method for electron dynamics of highly excited complex systems · PBE-lg and XYGJ-OS quantum mechanics methods for accurate intermolecular interactionsat modest cost ·The 2PT method for fast accurate calculations of entropy from molecular dynamics that we will illustrate with recent applications to Energy, Catalysis, Nanotechnology, and Materials selected from:

Applications of Multiparadigm, Multiscale Methodologies to high velocity impact ·EOS and phase transitions of materials from 2PT analysis of ReaxFF-lg reactive dynamics simulations Spin-coupling and superexchange in ferroelectrics ·Mechanism of superconductivity in cuprates; strategies for increased Tc ·Copper-Indium-Gallium-Selenide (CIGS)/CdS photovoltaics ·Hydration and hydrolysis processes in concrete (Ettringite) Functionalizing MOF, COF, ZIF Materials for storage and partitioning of H_2, CO_2, CH_4 ·Mechanisms of fuel cell catalysts including the oxygen reduction reaction ·Solid acid, Solid oxide, alkaline, and ceramic electrolytes ·simulations of transport in dye synthesized photovoltaics including electrodes, ionic liquid,and reluctant · New anodes and electrolytes for Li batteries · ReaxFF-Monte Carlo methods for resolving partial occupations from Rietveld analyses into supercells with whole atoms; application to Mixed metal oxide catalysts for ammoxidation of propane

Characterization of Saharan dust from red rain precipitated over Athens, Greece

A. GODELITSAS[1]*, P. NASTOS[1], T.J. MERTZIMEKIS[1], K. TOLI[1], A. DOUVALIS[2] AND R. SIMON[3]

[1]University of Athens, Greece (*correspondence: agodel@geol.uoa.gr)
[2]University of Ioannina, Greece
[3]ANKA Synchrotron Facility, KIT, Germany

Aeolian transport of Saharan dust influences significantly the rain acidity and furthermore the climate of the Mediterranean, causing among others, intense “red (or mud) rain” and even “red snow” episodes. During these episodes geological material from Sahara is deposited to the aquatic, terrestrial and urban environment [e.g. 1-3]. The Saharan dust samples were collected on membrane filters after intense “red rain” episodes over Athens megacity, Greece. Initial characterization by means of XRD, SEM-EDS and laser micro-Raman showed quartz, calcite and dolomite as major phases as well as phyllosilicates (mostly clays), rutile, zircon and goethite as minor constituents [4]. Preliminary analyses of metals using bulk XRF indicated Fe, Sr, Mn, Zn, Pb, Cr, Ni and Cu whereas gamma-ray spectroscopic measurements showed very low natural radioactivity and absence of human-produced nuclides. Detailed Synchrotron micro-XRF studies proved the presence of very hazardous elements, such as Pb and As, which had not been located on the samples by preliminary conventional investigation using SEM-EDS. It was also confirmed that many trace elements are intercorrelated (e.g. Fe-Mn-V-Cu) in other minor phases, of potential anthropogenic origin, hosted into the carbonate-silicate matrix. Subsequent Mössbauer spectroscopic study showed abundant Fe^{3+}-containing constituents and less Fe^{2+} phases. Moreover, sequential leaching experiments, using appropriate acids and ICP-MS analyses, revealed a high percentage of extractable Fe (and also Zn, Mn and Pb) due to carbonate phases comprising ~60% of the material. The above data can be important [see e.g. 5] for the geoavailability and bioavailability of aerosol-derived useful and harmful metals in southern Greece and generally in the entire semi-closed low-nutrient / low-chlorophyll marine ecosystem of eastern Mediterranean.

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