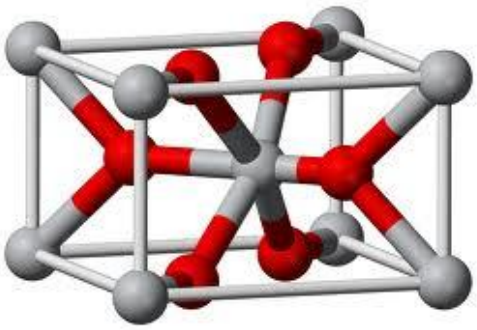
Thesisonderwerp 2012-2013 / **MM_12_NANO_14**

UNRAVELING THE PLASMA-CATALYTIC CONVERSION OF GREENHOUSE GASES WITH DENSITY FUNCTIONAL THEORY (DFT)

Keywords: Sustainable technologies, Density Functional Theory, molecular modeling, reactive force fields

Plasma catalysis has recently gained significant scientific interest, for instance for the destruction of VOC's and the conversion of greenhouse gases into value-added chemicals. On the fundamental level, however, the conversion mechanisms at the plasma-catalyst interface are virtually unknown. Current experimental investigations are based on a hit-or-miss approach, i.e. arbitrary changes of the plasma conditions are tested and their impact on the product yield of the catalytic process are measured. Atomistic insights, e.g. intermediate (short-lived) species at the surface and essential reaction mechanisms, are currently a missing link in the field of plasma catalysis. These atomistic insights are of fundamental importance to rationalize the design of a plasma catalyst.

Density Functional Theory (DFT) is nowadays the most attractive method to study chemical reactions at the nanoscale in computer simulations. This method is extensively used to study the individual steps in a reaction network, and will play a central role in this thesis. When one wants to study entire networks of concurrent chemical reactions at the nanoscale, the DFT method becomes computationally too demanding. Reactive force fields (e.g. ReaxFF) provide an alternative (and approximate) method to study chemical reactions at only a small fraction of the computational cost of DFT computations.



Goal

The conversion of greenhouse gases to value-added chemicals such as syngas and methanol on a anatase (TiO_2 polymorph) catalyst will be studied with DFT computations on periodic model systems with the VASP simulation program. The student will construct a database of reaction pathways between small molecules such as CO_2, H_2O, CH_4, CO, H_2, CH_3OH and related radical studies. These results will be used as a training set for the calibration of ReaxFF parameters in collaboration with the PLASMANT group (Prof. A.Bogaerts and Prof. K.Neyt, UA). The ALGC group of Prof. P.Geerlings and Prof. F.De Proft (VUB) has already analyzed the reactivity of greenhouse gases on gas-phase anatase model systems. Their experience and results can be used as a starting point for this master thesis.

The student will learn advanced simulation techniques currently used at the CMM to identify transition states of various catalyzed reactions at the anatase surface, such as the Nudge Elastic Band method.

Furthermore, new techniques to estimate and optimize reaction pathways, i.e. recently developed in the group of Prof. P.Ayers (McMaster University, Canada) will be tested in this work.

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