DFT investigations of PAHs in meteorites

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INTRODUCTION

The scope of this PhD project is to study the catalytic properties and activities of polycyclic aromatic hydrocarbons (PAHs), olivinic surfaces of meteorites and PAH-olivine complexes. We will support our computational approach with experimental investigations. It is already known that PAHs are formed in situ in the interstellar medium (ISM) and in solar system objects such as the Allende and Murchison meteorites, and they also are one of important sources for hydrogen production in ISM[1]. The PAHs present in meteorites predate the formation of these meteorites, but they could play an important role in the diversity of organic matter and on aqueous alteration of meteorites. Using quantum chemistry and in particular density functional theory (DFT)[5] we investigate the binding and barrier energies of PAHs such as pentacene, naphthalene, fluoranthene, coronene, hexabenzocoronene, interacting with olivinic surfaces ($(Mg,Fe)_2SiO_4$). We will study the effect of water ice and little molecules such as carbon dioxide and ammonia, in order to predict the potential energy surface (PES). Using this, we will derive a reasonable theoretical model with the help of different theoretical approaches. The ultimate goal of this project is to study the breakdown reaction of PAHs in order to understand the amino acid formation.

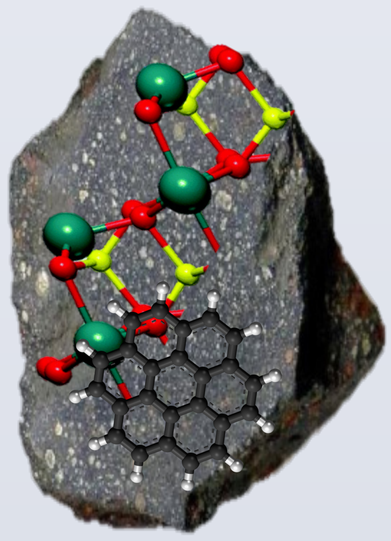
MAIN QUESTIONS

1) What role, if any, have minerals played in the diversity of organic matter in carbonaceous chondrites?

2) What role, if any, have PAHs[1,3] played in the organic inventory of meteorites?

3) How do the PAHs influence aqueous alteration of meteorites[4]?

4) Are PAHs indeed related to the formation of amino acids in meteorites[7]?



DENSITY FUNCTIONAL THEORY (DFT)

We will use DFT[5] to predict reaction paths and exotic species. By scanning the potential energy surface (PES) it is possible to estimate the barrier energy of the reaction, in order to understand the catalytic properties of the mineral and PAH surfaces. Furthermore, DFT allows to have accurate results with lower computational cost than other methods.

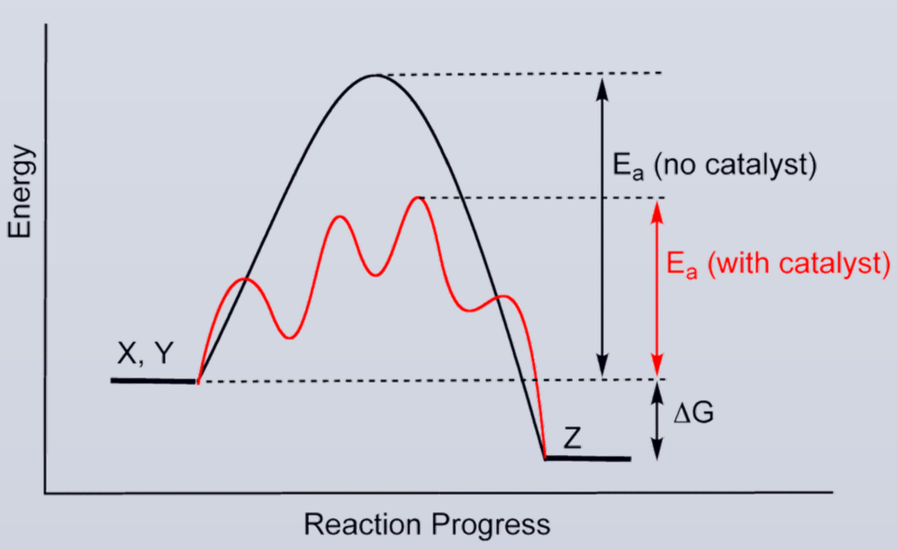


Figure 1
Potential Energy Surface (PES), showing reaction path with and without the presence of the catalyst.

BUILDING AN OLIVINE SURFACE

Periodic[8] and/or non-periodic approach[10]? which choice for this investigation?

DFT

Cluster Approach (QM, QM/MM)

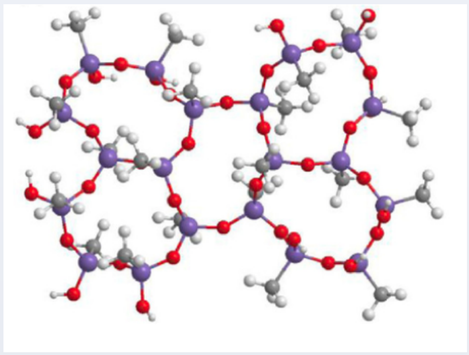


Figure 2
Molecular model representative of the cluster approach

Theory Level
M06-2X
pcseg-1

Periodic Approach

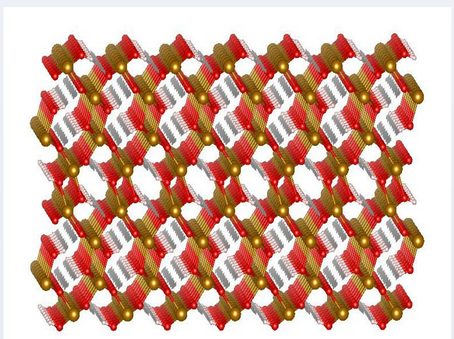


Figure 3
Molecular model representative of the periodic approach

Theory Level
PBE-D3, PAW,
500 eV ($E_{cut\ off}$)
K Points 7x3x7

In order to have accurate results it is necessary to start a calibration of computational methods (**Theory Level**) for testing the best functionals and basis sets[3] able to reproduce a determinate property.

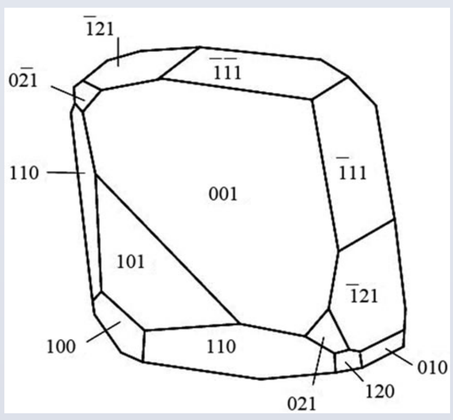


Figure 4
Crystal morphology.

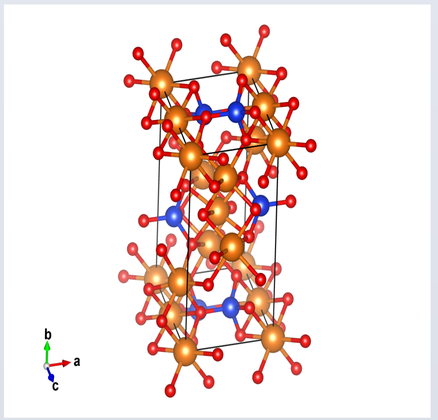


Figure 5
Unit cell of Forsterite. O in red, Si in blue and Mg in orange.

The structures, morphology, and stability of each Miller-index face (Figure 4) allow to understand which faces is more catalytic in order to model a proper surface related to the bulk property (Figure 5).

OUTLOOK AND NEXT STEPS

- **Systematic characterization of structures, composition and stabilities for dried, hydrated and hydroxylated surfaces of forsterite**
- **Serpentinization reaction due to the presence of PAHs.**
- **Interaction of PAHs on Forsterite surfaces (adsorption and dissociation)**
- **Interaction of PAHs on Forsterite surfaces in presence of point defects**
- **Investigation of the reaction path for the amino acids formation**

REFERENCES

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the extensive and ubiquitous role of polycyclic aromatic hydrocarbons in space

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