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New Technologies Based on Biological Catalysis

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Abstract – Efforts have been done to functionalize nanoporous and other nanostructures with enzymes to perform reactions massively with industrial interests. We have developed models of enzyme confinement in nanoporous and analyzed their conformational dynamics to understand how the nanocage environment affects the enzymatic activity. We also have studied biomembranes as biological catalyzers where reactants can be concentrated and orientated at membrane-water interfaces.

Biological molecular processes have become a rich source of inspiration for the development of advanced materials and new technologies. Natural selection imposes constant improvement, for example, in molecular recognition and in the catalysis of reactions at cellular level. Enzymes are between the most efficient catalyzers because they can convert reagents into products at rates above of 10^{10} times per second. Efforts have been done to functionalize nanoporous and other nanostructures with enzymes to perform reactions massively with industrial interests. A great challenge is to reproduce enzymatic activity in these materials with the same efficiency of the cells. It was observed that the confinement of the organophosphorous hydrolase metalloenzyme in functionalized mesoporous silica enhances the stability and increases catalytic specific activity by 200% compared to the enzyme in solution. However this is far below from cellular rates yet.

We have developed models of enzyme confinement in nanoporous (figure-1) to provide insights into how the nanocage environment steers enzyme conformational dynamics towards enhanced stability and enzymatic activity, and into how to improve them [1]. Comparisons of the RMSD curves and fluctuation amplitudes from the organophosphorous hydrolase MD simulations showed that the constraint of protein-nanoporous interface limits the phase space of the enzyme, reducing the number of conformations accessible to the catalytically competent form. The indiscriminate suppression of internal motions along the entire protein is likely to translate into decrease of catalytic efficiency. To prevent this problem in a more refined model, we bonded to the porous surface the functional group COO-(CH2)n, that is used experimentally, and we observed that the native fluctuations were restored and the configurational space can be better sampled for the catalysis efficiency.

The interactions of proteins with nanoporous are a key to the understanding of molecular confinement and interactive effects. This understanding is essential to direct the design and engineering of funcionalized enzyme complexes with a higher protein load capacity and improved biocatalyser performance. The nanoporous material, its dimension and geometry, and the substrates diffusion coefficients and collision rates also have very important roles.

Others biological catalyzers of great interest are biomembranes, where reactants can be concentrated and orientated at membrane-water interfaces. The improvement of reaction rates is due to the reduction of translational and rotational degrees of freedom of reactants. We have worked on membrane models in this sense also (figure-2). The reactions are simulated by hybrid methods of Molecular Mechanics and Quantum Mechanics (MM/QM), where the classical part is calculated using Gromos force field in the GROMACS package for Molecular Dynamics simulation, and the quantum part is calculated using RM1 Semiempirical Molecular Orbitals for enzyme systems and Carr-Parrinello Molecular Dynamics for membrane systems.

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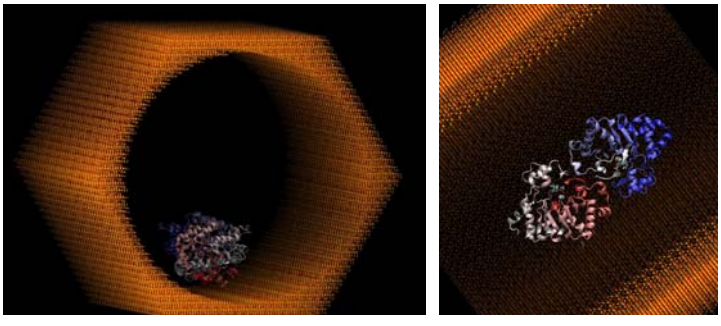

Figure 1: enzyme functionalized in nanoporous.

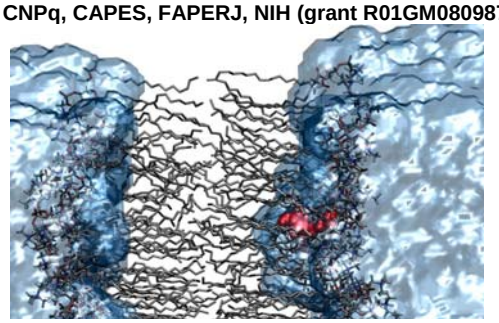

Figure 2: biomembrane (lipid chains), water (blue), and a reactant (red).

[1] D.E.B.Gomes, R.D.Lins, P.G.Pascutti, T.P.Straatsma, and T.A.Soares, Lect. Not. Comp. Science, 5167(2008) 68-78.