Valorisation at HIMS

Chemistry research that matters

Van ’t Hoff Institute for Molecular Sciences
The Computational Chemistry theme is leading worldwide in the fields of molecular simulations and multiscale modelling. Its aim is to develop computational tools to model and predict, from first principles, the behavior of complex chemical, biological, and physical processes.

Over the past decade the group has developed a strong alliance, the Amsterdam Center for Multiscale Modeling (ACMM), with its counterpart at the VU science faculty. The ACMM, established in 2007, has developed a strong High Performance Computing infrastructure and an internationally recognized training program.

The ACMM is world reference center in the field of research, training, and valorisation in the field of molecular multiscale modeling. Top research in all important modelling disciplines at one location, with direct access to essential infrastructure like the Supercomputer Center (SURFSARA) and the eScience Center.

Knowledge valorisation will also be facilitated via scientific consultancy for industry and the establishment of the ACMM-Laboratory (High Performance Computing infrastructure) that will be a hands-on hosting environment for commercial partners to learn and apply computational methods to systems of technological and industrial interest.

Molecular simulations
Biochemical and biophysical phenomena
Computational catalysis
Novel methodology development
Aqueous chemical processes
Nanostructured materials
Soft matter
Atomistic insight in biomolecular processes

**Challenge**
Understanding protein function requires knowledge of the structure, energetics and kinetics of the different intermediate states a protein can visit. Molecular simulation can provide exactly such knowledge, complementary to experiments. Molecular dynamics (MD) provides the necessary temporal and spatial resolution, as protein conformational changes are highly dynamical processes, in which thermal fluctuations play an important role. While MD in general has been hugely successful, addressing processes that take place on the millisecond to second time scale still poses a huge challenge.

**Solution:**
One way to overcome this challenge involves the use of effective bias potentials forcing the system to undergo the process of interest. Nevertheless, application of such potentials biases the outcome, especially in complex systems. Therefore, it is essential to obtain unbiased dynamics, which is possible by using transition path sampling, a computational framework that harvests MD trajectories that undergo reactions of interest.

**Mechanism of photoreceptor function**
Photoreactive Yellow Protein is a bacterial blue-light receptor. Blue light triggers a cascade of rearrangements in the protein.

**Protein DNA binding**
In bacteria, the Histone-like Nucleoid Structuring protein forms bridges between strands of duplex DNA.

**Amyloid fibril formation**
Aggregation of the insulin derived LVEALY peptide occurs via the lock-dock mechanism.

**Kinetics and mechanisms of protein folding**
The Tip-cage miniprotein is a model system for protein folding. We elucidated the mechanism and kinetics of the folding and unfolding of this small protein, using advanced simulation methods.

**Valorization goals**
Unravel kinetics and reaction mechanisms in biological processes
Guide and assist in the interpretation of experiments
Develop new efficient computational tools for the community
Understanding soft matter

**Challenge**

Soft materials such as colloids, emulsions, polymers, and surfactants, can have exceptional mechanical, optical or functional properties that find applications in both industry and society. Examples are found in consumer products such as shampoo, shaving cream, paint, plastics and food, but also in drug delivery systems. Soft matter easily deforms under external forces because forces and interactions act on mesoscopic scales. The components often self-organize into complex structures with striking mechanical, or functional properties. The key question is: How can we understand their structural, mechanical and (physico-) chemical properties from the building blocks and their interactions? Together with experimental groups we attack this problem using advanced molecular simulation methods.

**Self-assembly of polymer networks**

Telechelic polymers form complex networks depending on the functionality of the end-group. Asymmetric telechelic polymers can be triggered separately. Together with WUR researchers we investigate dependence on the order of the trigger sequence on the final network properties.

**Anisotropic self-assembly of colloidal particles**

Isotropic particles can self-assemble into anisotropic structures. Acting as nanofiller in polymer nanocomposites, lead to special mechanical properties.

**Predicting reaction coordinates of crystal nucleation**

We predict the poorly understood structural nature of the critical nucleus and the involved reaction coordinates, and explain the observed kinetically favored meta stable crystal phases.

**Active matter**

Active matter is a class of soft matter in which self propulsion plays an important role.

Examples of active matter on different scales: birds, fish, bacteria, people.

We investigated in dense colloidal suspension (81% volume fraction) how activity enhances crystallization and suppresses the glass transition.

We develop dynamics of microscopic filaments in fluids (micro-fluidics). In a circularly polarized field a initially misaligned fiber will align along the filed axis in a spiral motion.

**Valorization goals**

Understand and predict soft matter self-assembly processes

Guide and assist in the interpretation of experiments and development of devices

Provide control over material properties
Molecular Simulation
- DFT (Ab Initio) Interactions
- Empirical Force Fields
- Statistical Thermodynamics
- (Ab Initio) Molecular Dynamics
- Monte Carlo Methods
- Rare Event Sampling Methods

Solvent Effects in Chemical Reactions
- Transfer Hydrogenation
  - Metal-Catalyzed Transfer Hydrogenation
  - Important Factors:
    - Ligands
    - Nature of Solvent
    - Proton transfer
  - Ab Initio Model
  - Reaction Free Energy
  - Reactive Pathways of Solvent Mediated Mechanism
- Silica Oligomerization
  - First Step in Zeolite Synthesis
  - Important Factors:
    - pH of solution
    - Structure Directing Agents (Ions)
    - Kinetic Network
- Aqueous Solutions
  - Important Factors:
    - Ionic Size
    - Structure Making/Breaking
    - Proton Mobility
    - Ions Co-Factor in Aqueous Chemistry
  - IR Spectra
  - Ionic Hydration and Protons Mobility

ValORIZATION GOALS
- Screening of Compounds and Materials
- Predictive Modeling for a Wide Range of Conditions
- Rational Design of Novel Processes and Compounds

References
Molecular Simulation

DFT (Ab Initio) Interactions
Empirical Force Fields
Statistical Thermodynamics

(Ab Initio) Molecular Dynamics
Monte Carlo Methods
Rare Event Sampling Methods

Multiscale Modeling

ACMM

High Accuracy Method Development
MD, DFT

Fluid Dynamic Reactors

Molecular Simulation

MOFs
Important Factors:
Size selectivity (sieving)
Shape Selectivity
Packing effects
Preferential Interactions

Xylenes Separation using MOFs

Xylenes in MAF-X8
Commensurate Stacking

Xylenes in MAF-X8
Selective Adsorption

Xylenes in MAF-X8
Breakthrough Profiles

Xylene Loading
MOFs Compared

Nanoporous Materials and Surfaces

Van 't Hoff Institute for Molecular Sciences

Computational Chemistry

Molecular Simulation

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Methanol to Olefin Conversion In Zeolites

MTO is Acid Catalyzed Process
Important Factors:
Proton Mobility - MeOH/H2O Ratio and Loading

Proton trajectories

5 MeOH / acid site
8 H2O / acid site

Clay/Water Interfaces

Important Factors:
Cat-/Anions Adsorption - Surface Hydration - Acidity Surface Groups - Chemical Reactivity

Water / Proton / Hydroxyl Association and Dissociation

Aluminium Oxides/Water Interfaces

Aluminium Oxides Important Heterogeneous Catalyst
Crucial Factors for Reactivity:
Surface hydration - Doping - Acidity/Basicty Surface Sites

Hydroxyl Types on Alumina
AIMD of Alumina/Water Interface

ValORIZATION GOALS
- Screening of Compounds and Materials
- Predictive Modeling
- Rational Design and Engineering with Atomistic Precision
- Software Suite for Nanoporous Materials
- Patents

References
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  Dubbeldam., Angew. Chem. Int.
  Ed. 53, 7774 (2014).
- X. Liu, X. Lu, M. Sprik, J. Cheng,
  E.J. Meijer, R. Wang, Geo, Cosm.
Multiscale Modelling of Complex Materials

By combining Electronic Structure Calculations with Molecular Dynamics Simulations, we unravel complex molecular phenomena in catalysis, biophysics, and material science. In-house developed simulation methods allow us to study larger molecular systems and longer time-scales. We use advanced sampling techniques to probe activated transitions and reaction dynamics.

Polymer dynamics

Enzyme catalysis

Bio-photo-sensing

Spectroscopy interpretation

Redox properties electron transfer

Acidity / proton transfer

Electronic structure

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Electronic structure

Large polymer and biomolecular systems are simulated with forcefield based MD or the hybrid QM(DFT)/MM and coarse-grain/atomistic methods. Where necessary, forcefields are fitted against accurate electronic structure calculations. Development of theory, algorithms and computer code, allows us to calculate specific properties and observables that are not available in commercial modelling programs.

Proton and electron transfer processes are simulated with DFT-MD in different molecular environments to compute for example conductivity, pKa, and redox potentials.

The free energy landscape gives direct insight in the reaction mechanisms and reaction rates. With our metadynamics simulations, we probe catalytic reactions in solution, at interfaces, and in biomolecules.

References

Our multiscale modeling approach is widely applicable to:
- Unravel and optimize reaction mechanisms
- Predict structure and dynamics of molecular systems
- Interpret experimental spectra and measurements
Gelation in crosslinking polymerization: multiple radical sites that matter

Crosslinking polymerization, that is known to produces polymers with complicated branched topology due to crosslinking reaction mechanism:

$$R_{1} + R_{2} \rightarrow R_{1} + R_{2}$$

has been studied by means of a four-dimensional population balance model accounting for chain length $x$, free pending double bonds $y$, crosslinks $c$, and radicals $z$ as dimensions. The model, for the first time and to a full extent resolves the crosslinking problem as formulated by Shiping Zhu two decades ago, and covers both pre-gel and gel regimes in a straightforward manner.

The model has been validated with data from an experimental crosslinking polymerization, Methyl Methacrylate with Ethylene Glycol Dimethacrylate. Non-trivial patterns in the time evolution of average quantities like crosslink densities, partly observed in prior studies, are naturally emerging from the model by computing marginal of the four-dimensional distribution possessing an interesting multimodal structure.

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*References*

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- Polymer 55(16), 3476–3489, 2014
- MTS 23, 7-14, 2014
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*Valueization goals*

Prediction of the topologies of branched polymer architectures and segment lengths from kinetics.