UNIVERSITY OF AMSTERDAM

Valorisation at HIMS

Chemistry research that matters

Van 't Hoff Institute for Molecular Sciences

2015

University of Amsterdam

Computational Chemistry

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

ACMM Amsterdam Center for Multiscale Modeling

Universiteit van Amsterdam — vrije Universiteit amsterdam

Computational Chemistry

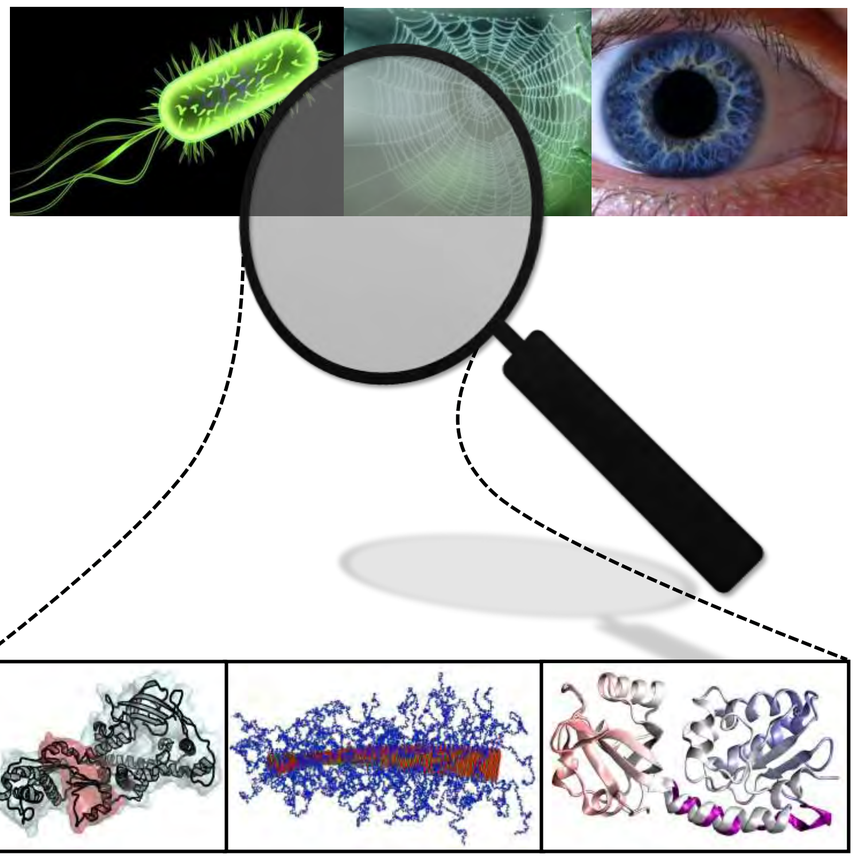
Atomistic insight in biomolecular processes

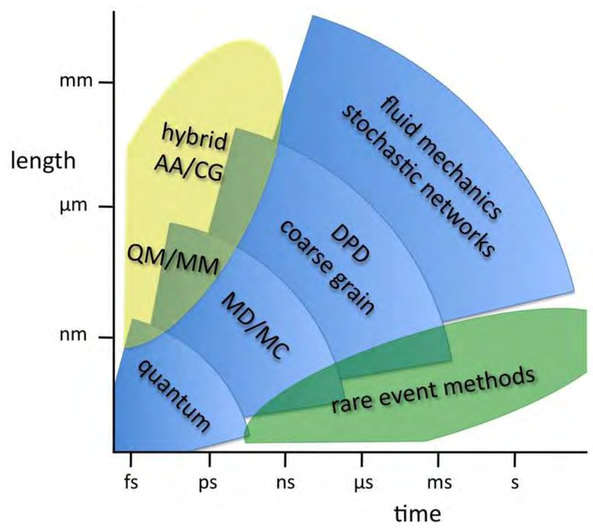
Challenge

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:

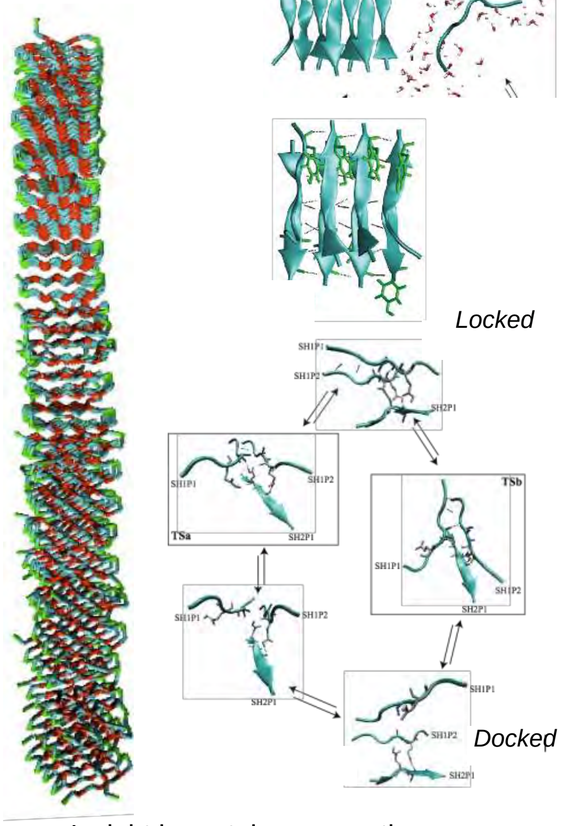
One way to overcome this challenge involves the use of effective bias potentials forcing the system to undergo the process of interest. Nevertheless, application of such potentials biases the outcome, especially in complex systems. Therefore, it is essential to obtain unbiased dynamics, which is possible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





Amyloid fibril formation

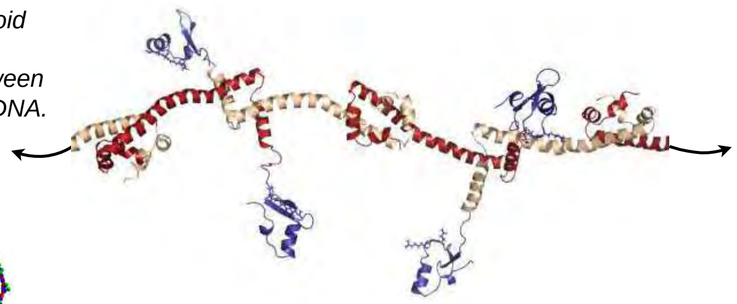
Aggregation of the insulin derived LVEALYL peptide occurs via the lock-dock mechanism.

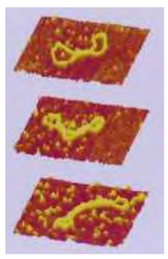


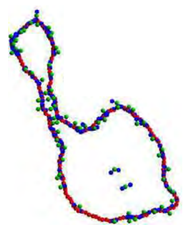
Insight in protein aggregation
Role of water; control of growth

Protein DNA binding

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





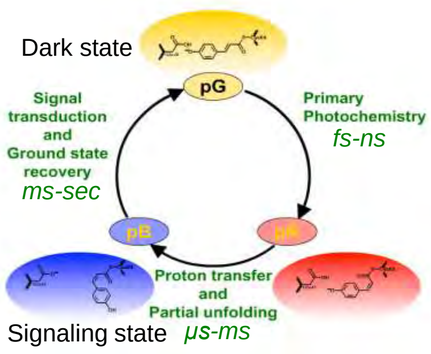


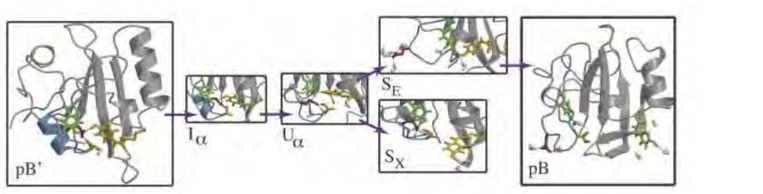
We have studied the DNA binding of H-NS at different length and time scales, resulting in the prediction of the mechanism of nucleotide sequence recognition and bridge formation.

Prediction of structure and recognition mechanism
Interpret and guide experiments

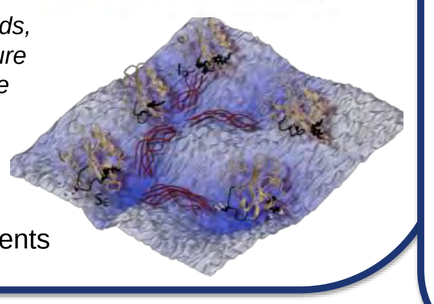
Mechanism of photoreceptor function

Photoactive Yellow Protein is a bacterial blue-light receptor. Blue light triggers a cascade of rearrangements in the protein.



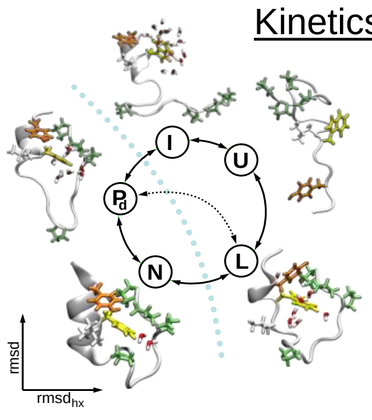


Using advanced simulation methods, we were able to predict the structure and mechanism of formation of the signaling state.

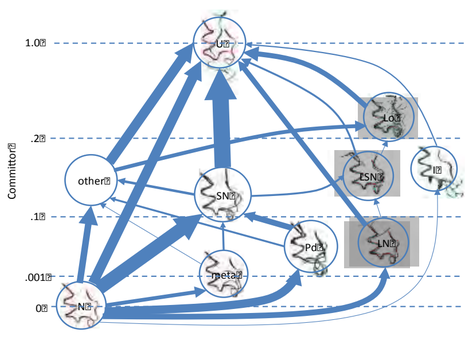


Prediction of structure and mechanism
Interpret and guide experiments

Kinetics and mechanisms of protein folding



The Trp-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.



Development of simulation tools for studying kinetics of rare events such as protein folding

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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

ACMM Amsterdam Center for Multiscale Modeling

Universiteit van Amsterdam — vrije Universiteit amsterdam

Computational Chemistry

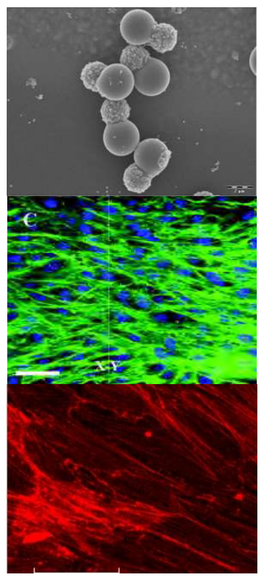
Understanding soft matter

Challenge

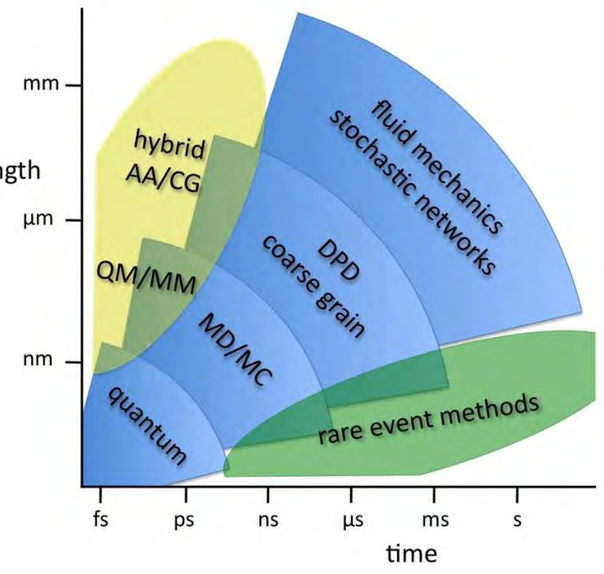
Soft materials such as colloids, emulsions, polymers, and surfactants, can have exceptional mechanical, optical or functional properties that find applications in both industry and society. Examples are found in consumer products such as shampoo, shaving cream, paint, plastics and food, but also in drug 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?

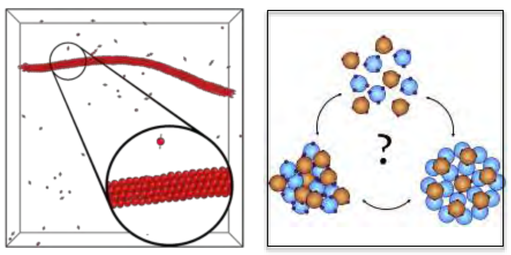
Together with experimental groups we attack this problem using advanced molecular simulation methods.



Interpretation
Guidance



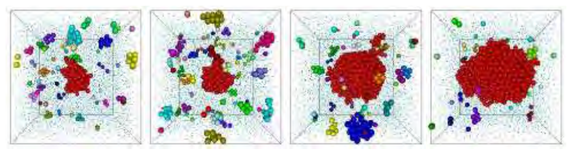
Inspiration



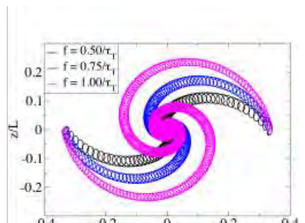
Active matter

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 (61% volume fraction) how activity enhances crystallization and suppresses the glass transition.

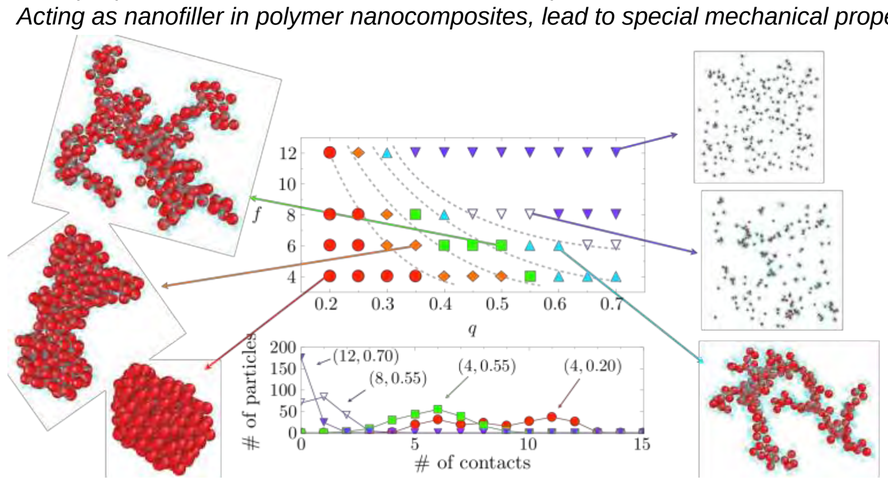


We develop dynamics of microscopic filaments in fluids (micro-fluidics). In a circularly polarized field a initially misaligned fiber will align along the filed axis in a spiral motion.

Understanding of active matter properties

Anisotropic self-assembly of colloidal particles

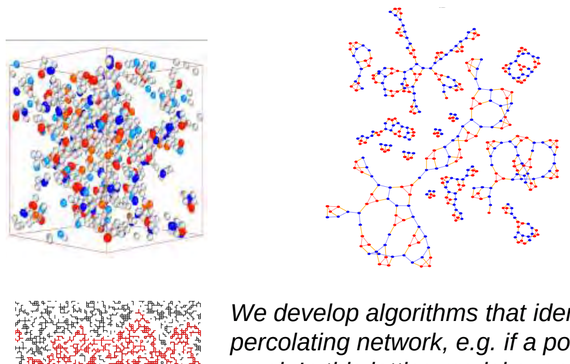
Isotropic particles can self-assemble into anisotropic structures.
Acting as nanofiller in polymer nanocomposites, lead to special mechanical properties



Understanding and prediction of colloidal self assembly processes.
Interpret and guide experiments

Self-assembly of polymer networks

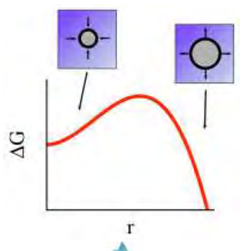
Telechelic polymers form complex networks depending on the functionality of the end-group. Asymmetric telechelic polymers can be triggered separately. Together with WUR researchers we investigate dependence on the order of the trigger sequence on the final network properties.

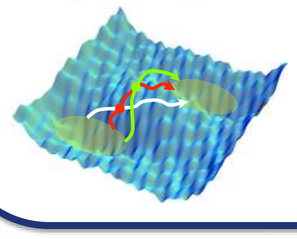


We develop algorithms that identify a percolating network, e.g. if a polymer forms a gel. In this lattice model our algorithm proves the largest connected grouping (red) fails to connect over all space when repeated periodically. If just the points marked blue were connected it would.

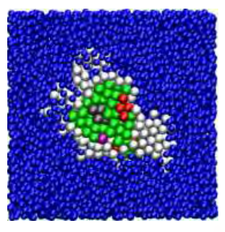
Prediction of structure and mechanism
Interpret and guide experiments
Development of tools for analysis of networks

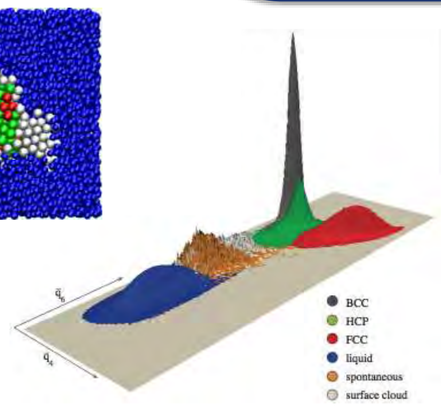
Predicting reaction coordinates of crystal nucleation





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.





Development of simulation tools for studying kinetics of rare events such as crystal nucleation

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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

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Computational Chemistry

RESEARCH PRIORITY AREA SUSTAINABLE CHEMISTRY

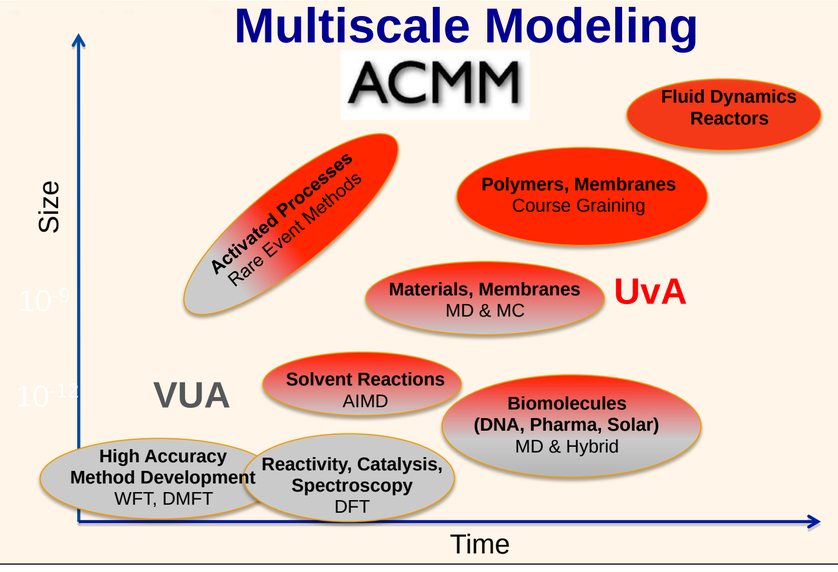
Van 't Hoff Institute for Molecular Sciences

Molecular Simulation

DFT (Ab Initio) Interactions
Empirical Force Fields
Statistical Thermodynamics

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

Multiscale Modeling

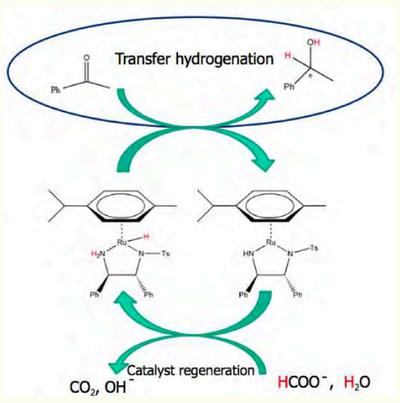


Solvent Effects in Chemical Reactions

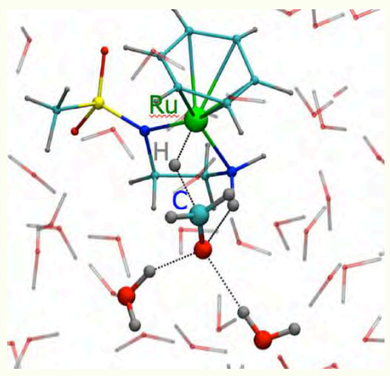
Transfer Hydrogenation

Metal-Catalyzed
Transfer Hydrogenation

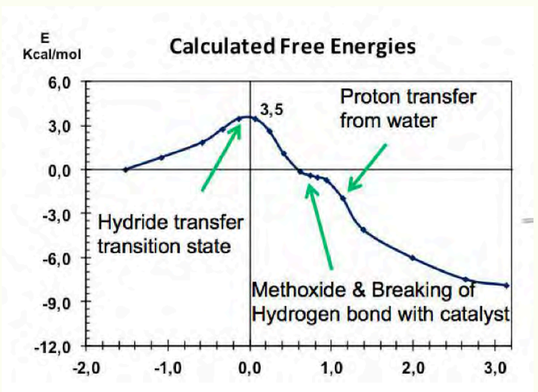
Important Factors:
Ligands
Nature of Solvent
Proton transfer



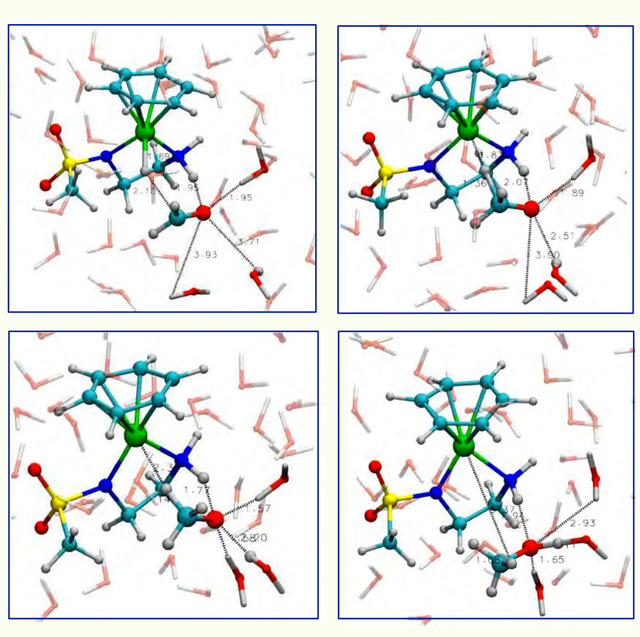
Ab Initio Model



Reaction Free Energy



Reactive Pathways of Solvent Mediated Mechanism

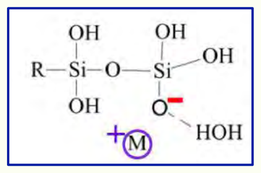


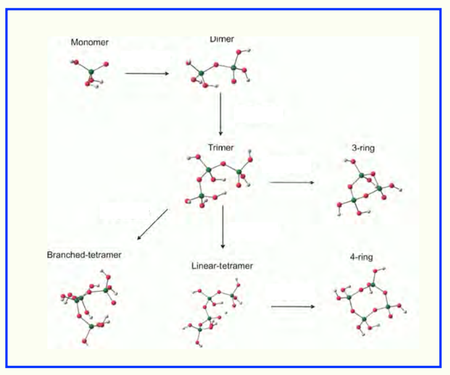
Silica Oligomerization

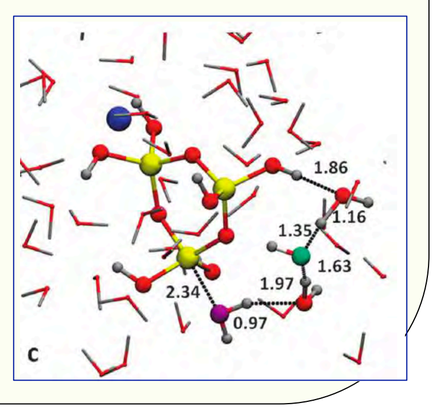
First Step in Zeolite Synthesis

Important Factors:
pH of solution
Structure Directing Agents (Ions)
Kinetic Network

Reaction Mechanism in Solution: Water and Ion Effects



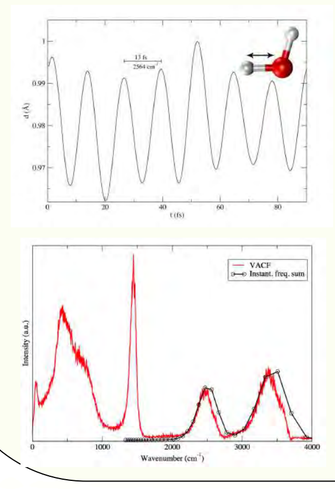




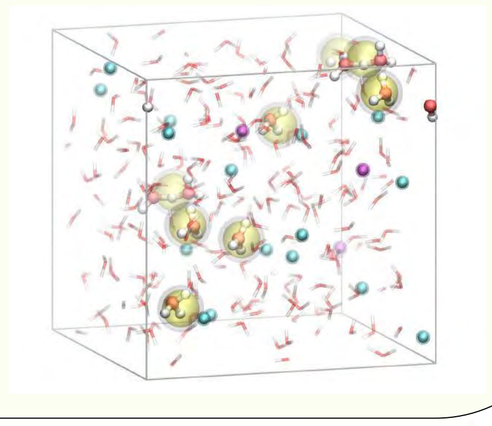
Aqueous Solutions

Important Factors
Ionic Size – Structure Making/Breaking – Proton Mobility
Ions Co-Factor in Aqueous Chemistry

IR Spectra



Ionic Hydration and Protons Mobility HCl/MgCl Solution



Molecular Simulation

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References

- A. Pavlova and E.J. Meijer, Chem. Phys. Chem. 13, 3492 (2012)
- A. Pavlova, T.T. Trinh, R. van Santen, E.J. Meijer, Phys.Chem.Chem.Phys. 15, 1123 (2013).

Valorization goals

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

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Computational Chemistry

RESEARCH PRIORITY AREA SUSTAINABLE CHEMISTRY

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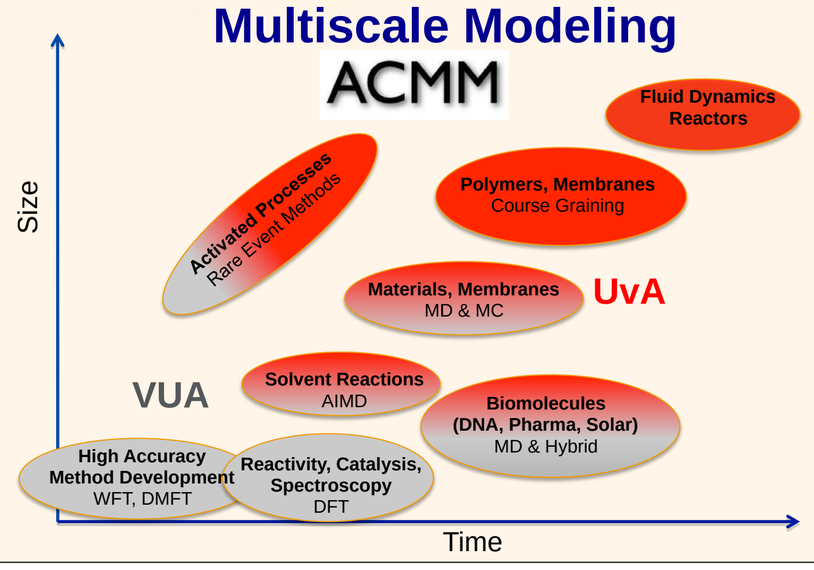
Multiscale Modeling

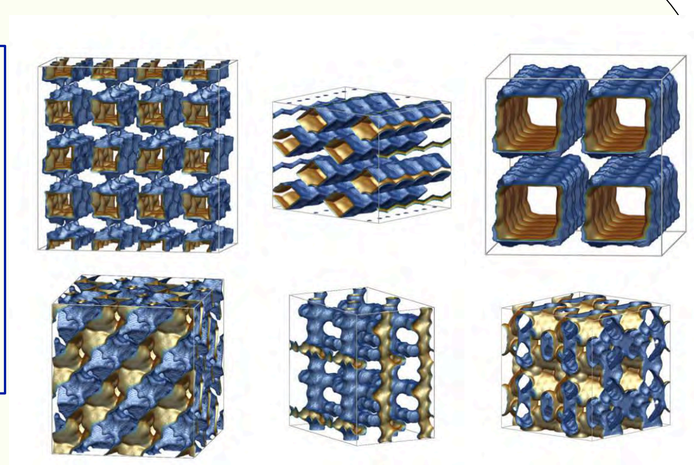


Nanoporous Materials and Surfaces

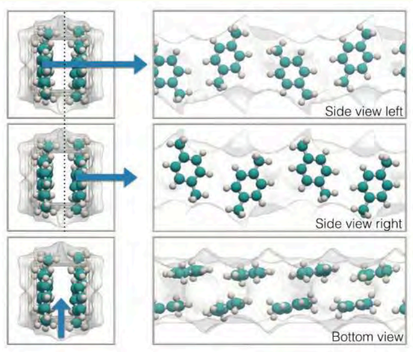
Xylenes Separation using MOFs

MOFs

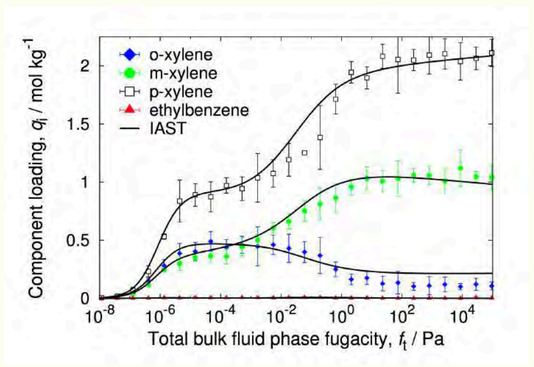
Important Factors:
Size selectivity (sieving)
Shape Selectivity
Packing effects
Preferential Interactions



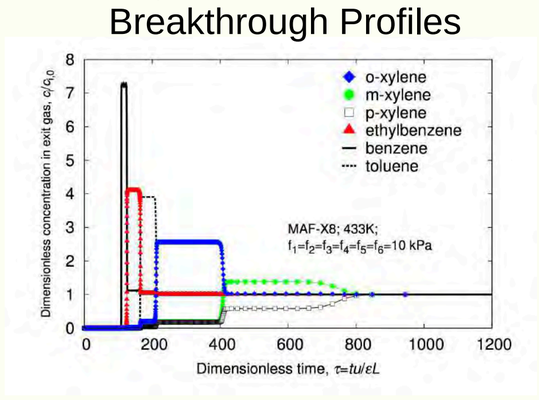
Xylenes in MAF-X8
Commensurate Stacking



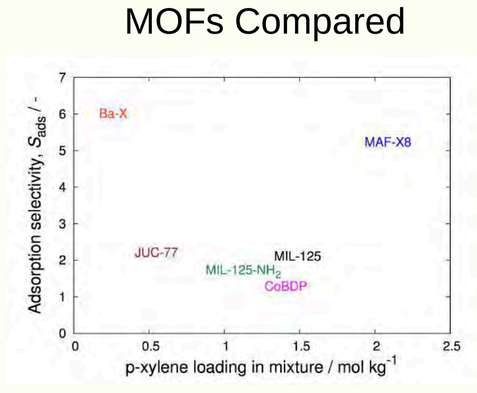
Xylenes in MAF-X8
Selective Adsorption



Xylenes in MAF-X8
Breakthrough Profiles



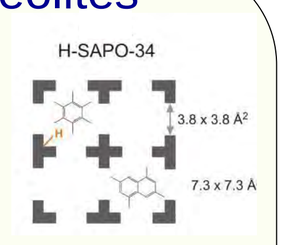
Xylene Loading
MOFs Compared



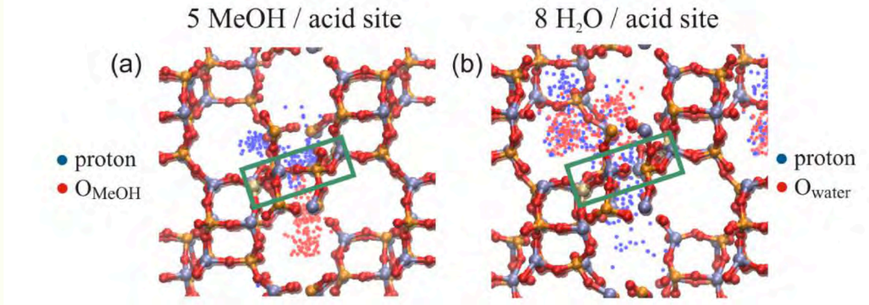
Methanol to Olefin Conversion In Zeolites

MTO is Acid Catalyzed Process

Important Factors:
Proton Mobility - MeOH/H2O Ratio and Loading

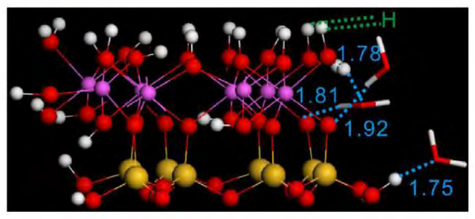


Proton trajectories



Clay/Water Interfaces

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



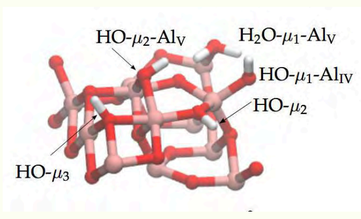
Water / Proton / Hydroxyl
Association and Dissociation

Aluminium Oxides/Water Interfaces

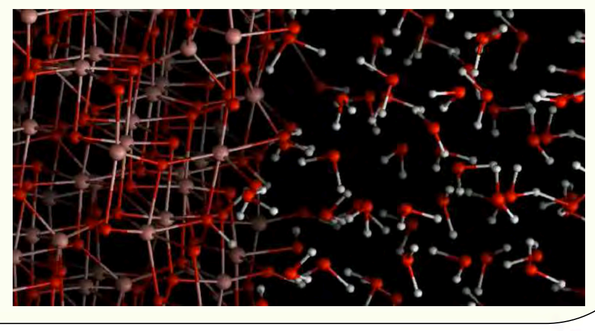
Aluminium Oxides Important Heterogeneous Catalyst

Crucial Factors for Reactivity:
Surface hydration - Doping - Acidity/Basicity Surface Sites

Hydroxyl Types on Alumina



AIMD of Alumina/Water Interface



Molecular Simulation

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- X. Liu, X. Lu, M. Sprik, J. Cheng, E.J. Meijer, R. Wang, Geo. Cosm. Acta. **117**, 180 (2013).

Valorization goals

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

ACMM Amsterdam Center for Multiscale Modeling

Universiteit van Amsterdam · vrije Universiteit amsterdam

Computational Chemistry

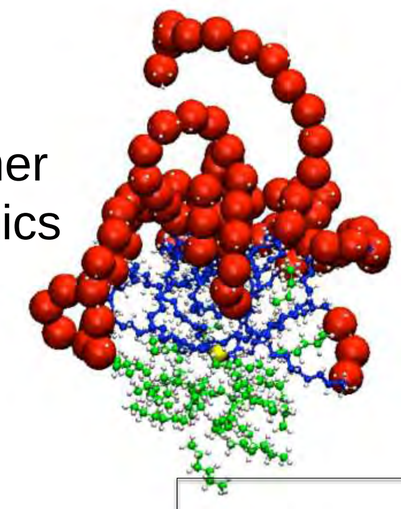
RESEARCH PRIORITY AREA SUSTAINABLE CHEMISTRY

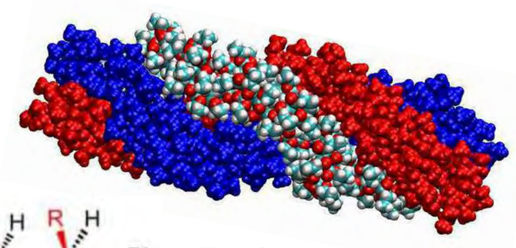
Multiscale Modelling of Complex Materials

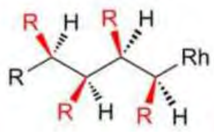
By combining Electronic Structure Calculations with Molecular Dynamics Simulations, we unravel complex molecular phenomena in catalysis, bio-chemistry, and material science.

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.

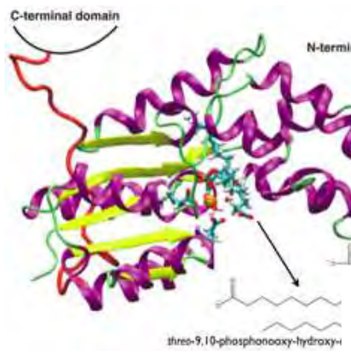
Polymer dynamics



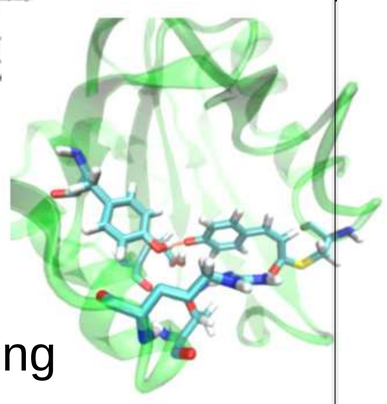




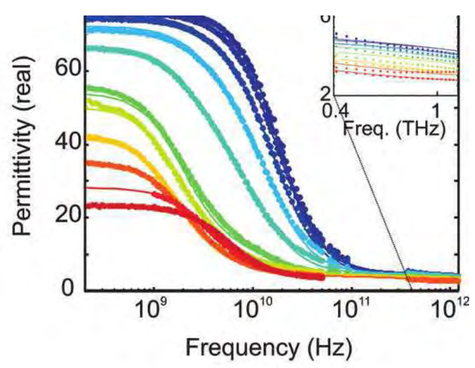
Polymer structure



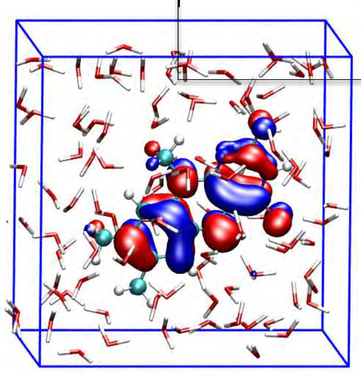
Enzyme catalysis



Bio-photo-sensing

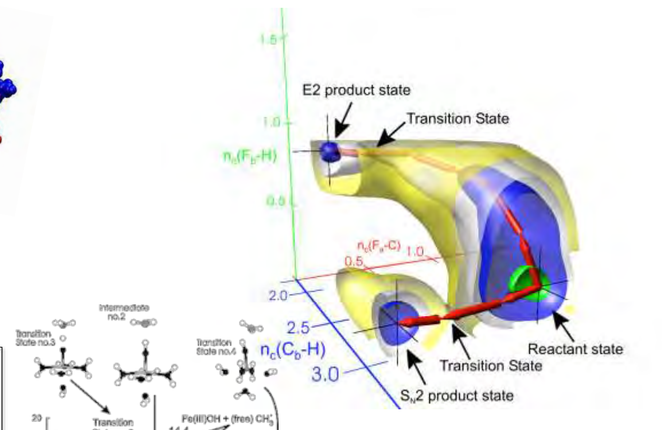


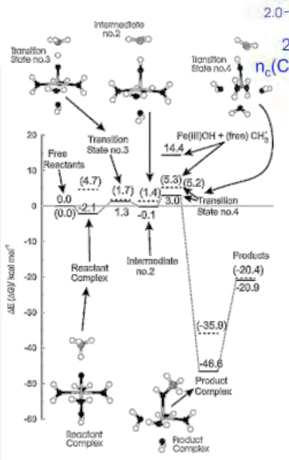
Spectroscopy interpretation



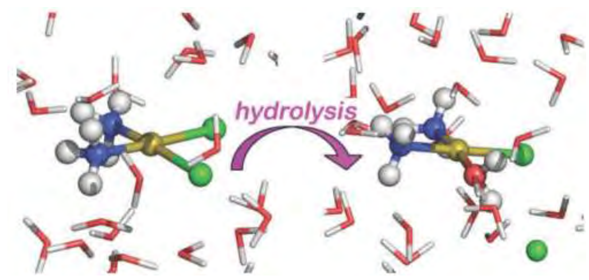
Redox properties

electron transfer

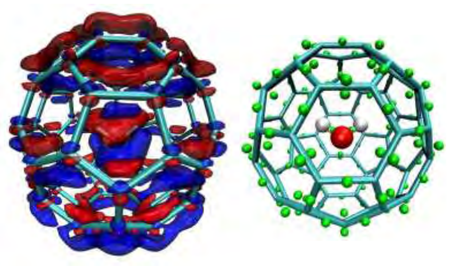




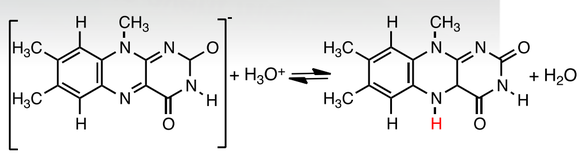
Reaction mechanics & dynamics



Electronic structure



Acidity / proton transfer



Large polymer and biomolecular systems are simulated with **forcefield based MD** or the **hybrid QM(DFT)/MM** and **coarse-grain/atomistic** methods. Where necesary, 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.

Computational Chemistry

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URL: www.acmm.nl

References

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Phys. Chem. Chem. Phys. 16 (2014), 18993

Valorization goals

Our multiscale modeling approach is widely applicable to:

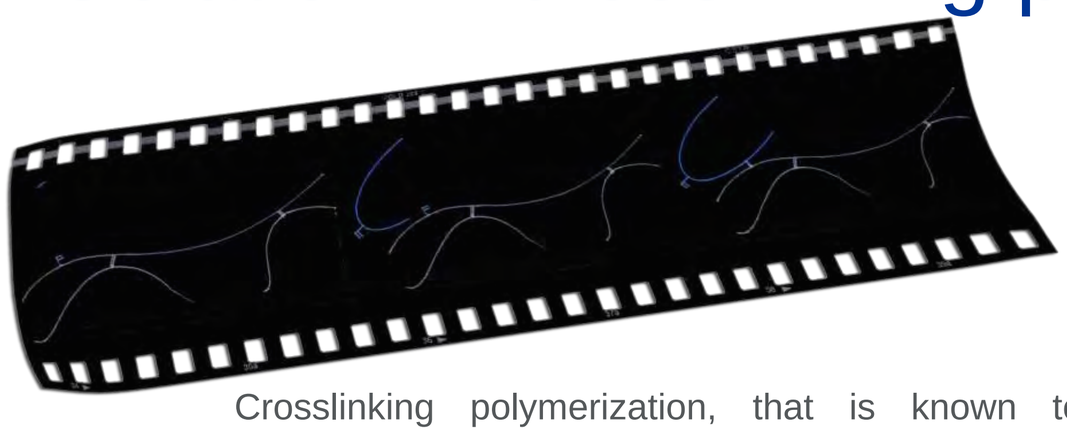
- Unravel and optimize reaction mechanisms
- Predict structure and dynamics of molecular systems
- Interpret experimental spectra and measurements

University of Amsterdam

Computational Chemistry

Van 't Hoff Institute for Molecular Sciences

Gelation in crosslinking polymerization: multiple radical sites that matter



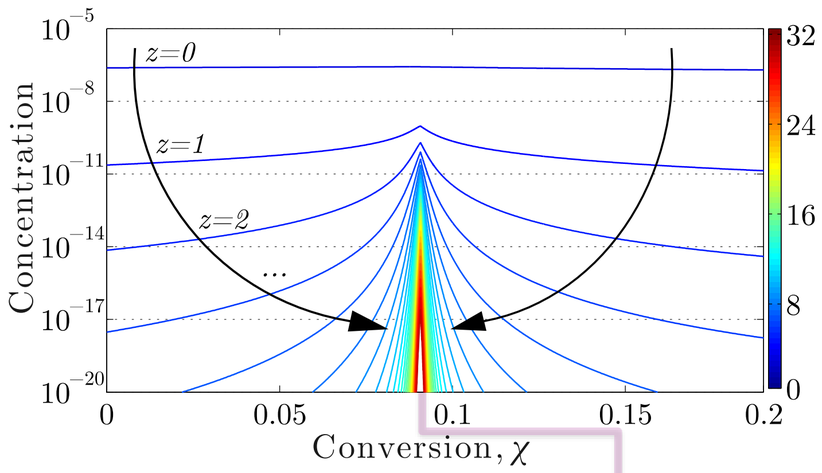
Crosslinking polymerization, that is known to produces polymers with complicated branched topology due to crosslinking reaction mechanism:

$$R_{x_1,y_1,c_1,z_1} + R_{x_2,y_2,c_2,z_2} \xrightarrow{y_1 z_2 k_c} R_{x_1+x_2,y_1+y_2-1,c_1+c_2+1,z_1+z_2}$$

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.

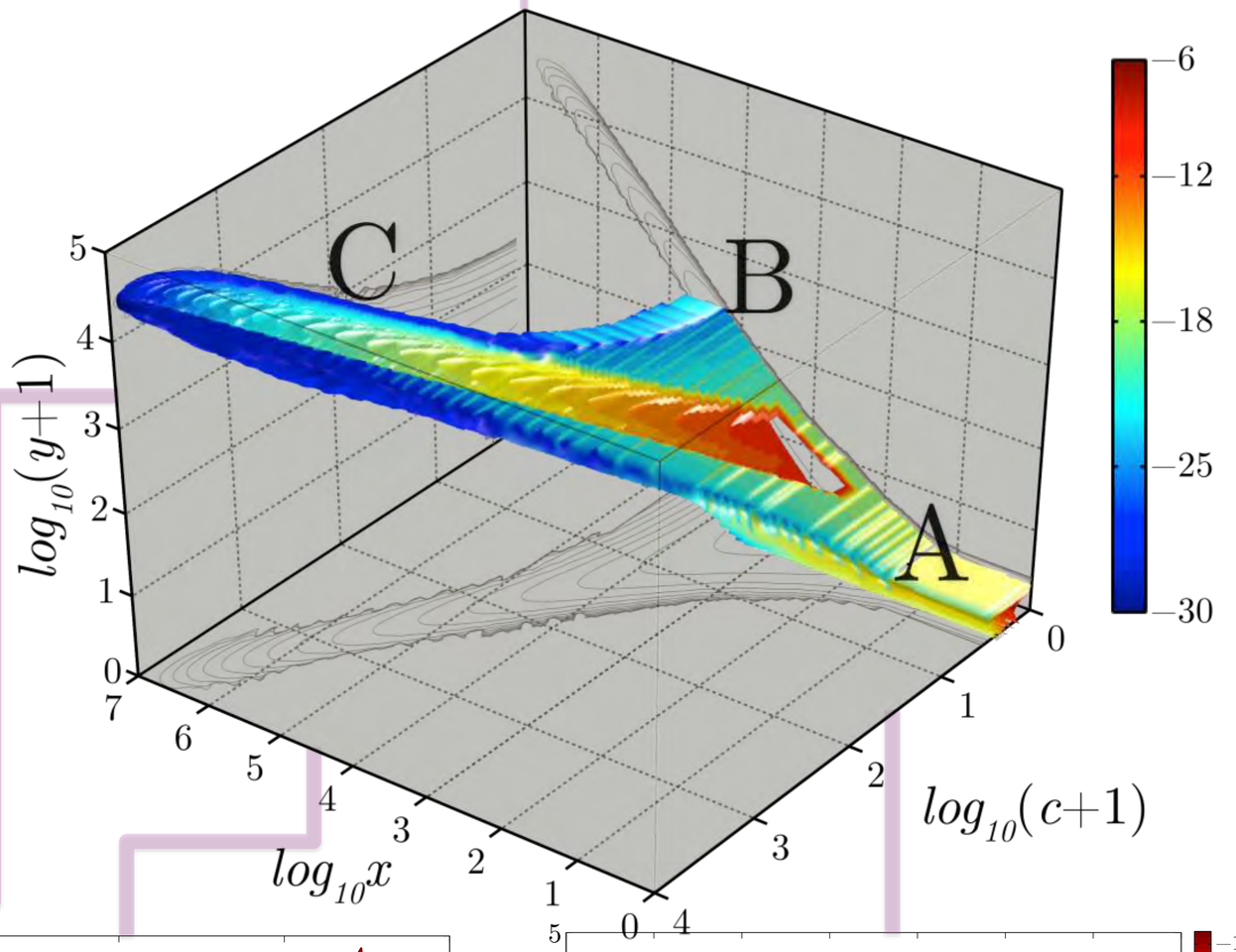
The model has been validated with data from an experimental crosslinking polymerization, Methyl Methacrylate with Ethylene Glycol Dimethacrylate. Non-trivial patterns in the time evolution of average quantities like crosslink densities, partly observed in prior studies, are naturally emerging from the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

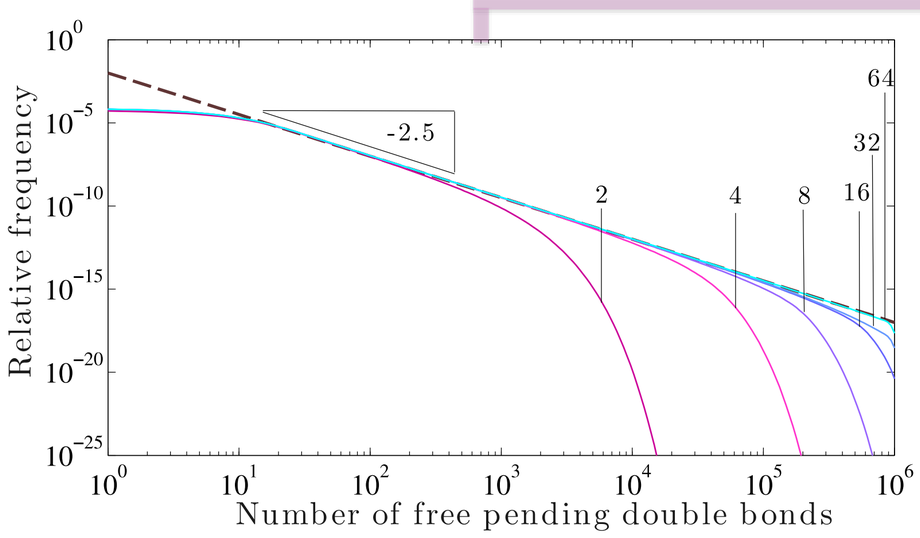
The work described here was financed by the DPI



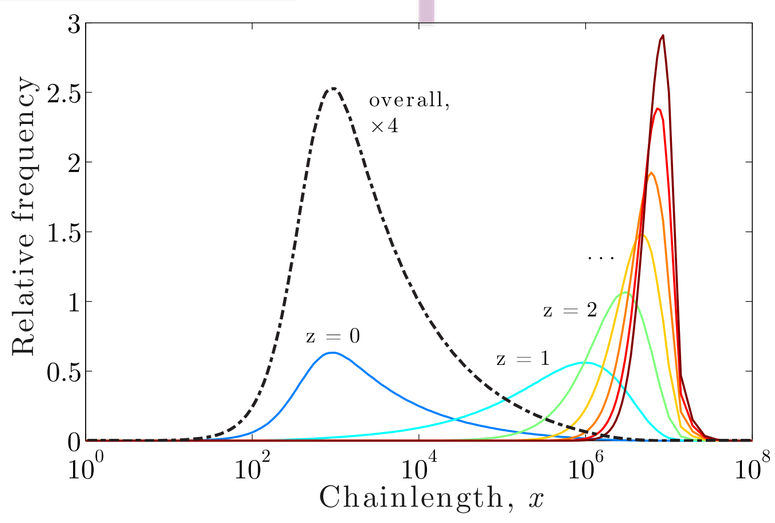
Evolution of multiradicals

Concentration of molecules with various numbers of radical sites as the reaction passes the gelation point.
A significant number of multiradicals is present temporary around the gel point.

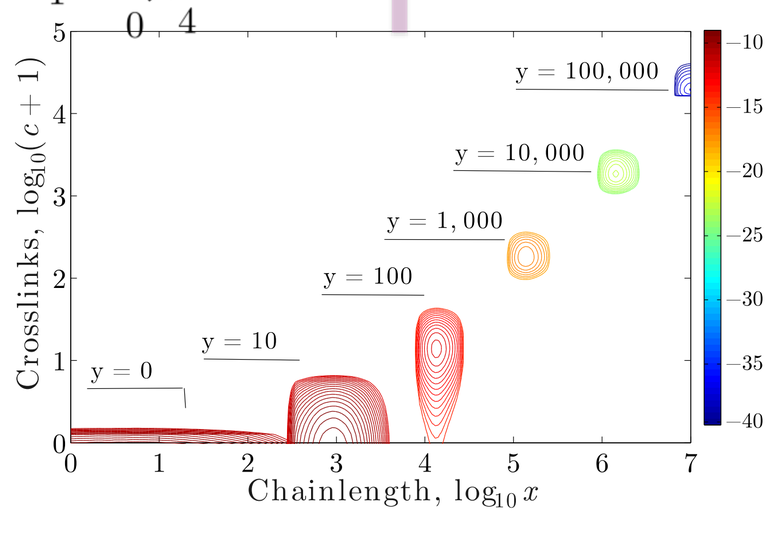




FPDB distribution obtained from models with different maximum number of radical sites per molecule. The dashed line depicts an asymptote of the tail of an FPDB distribution with no restrictions on radical sites number: an algebraic decay proportional to $x^{-2.5}$



GPC chain length distributions at the gel point obtained in each radical class *z* are shown as solid lines. The values of the overall distribution, depicted by a dashed line, are scaled by a factor 4 for comparison



Sol molecules can be separated according to number of FPDB they posses. Chain length/crosslinks distributions for classes of molecules with a fixed number of FPDB, emerge as narrow peaks

Computational Polymer Chemistry

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References

I. Kryven *et al* in:

- Polymer 55(16), 3475–3489, 2014
- MTS 23, 7-14, 2014
- Polymer, 54(14), 3472–3484, 2013
- MRE 7 (5), 205-220, 2013

Valorization goals

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