Multiscale Simulations of Polymers

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Polymer phenomena and their relevance

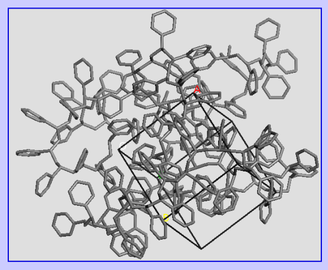
So, what is a polymer and why do we need to simulate it at all? A polymer is a large molecule, often containing many thousands of small monomers joined together chemically to form one giant macromolecule. Polymers have their own characteristic phenomena such as very complex structure, slow relaxation, many length and time scales. Fundamental insight generated by multiscale computer simulations will help in developing new materials with improved ultimate properties. In our research the major emphasis is on atomic-scale modeling of macromolecules, including glasses, dendrimers, polymer nanocomposites and thin polymer films. The focus is – obviously! – on polymer dynamics and mechanics.

Multiscale Approach

The mechanical properties of amorphous polymers are intimately linked to the collective segmental dynamics of individual polymer chains. The existent multi-scale modeling approach to mechanical response of polymers still lacks a connection with the molecular level. Our work involves large scale computer simulations of realistic polymer models using atomistic molecular dynamics (MD), Monte Carlo (MC) and Brownian dynamics (BD) simulation techniques.

Molecular Dynamics: Atomistic Polymer Models

In MD, successive configurations of the system are generated by integrating classical Newton's equations of motion. Our goal here is to understand the link between the molecular structure of amorphous polymers in a glassy state and their bulk mechanical properties.

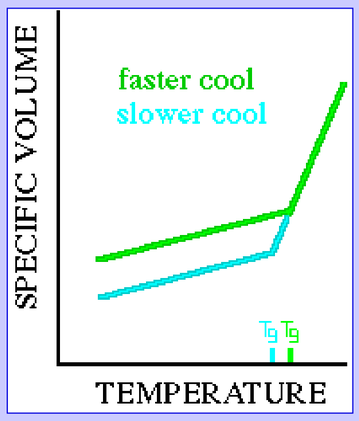


Conformation of a single polystyrene chain in a glassy state in a simulation box.

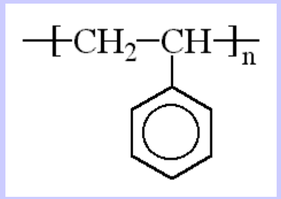
Glassy Polymer Dynamics

The glassy state of matter and the glass transition itself are still great, unsolved problems in condensed matter physics.

There is a certain temperature (different for each polymer) called the *glass transition temperature*, or *Tg* for short. Below this temperature the large-scale motions are practically frozen. Hard plastics like *polystyrene* are used below their glass transition temperatures.



Uses: thermoplastics
T_g: 100 °C
Monomer: styrene

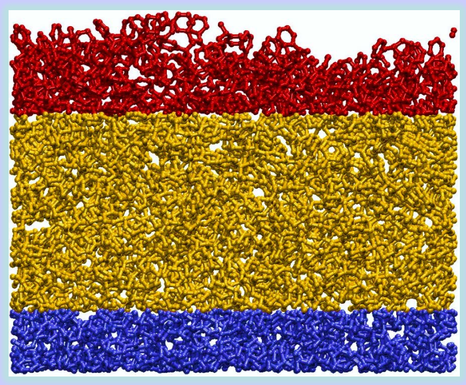


We simulate this transition using MD and try to understand the specifics of polymer glassy dynamics.

Thin Films: Confined Polymers

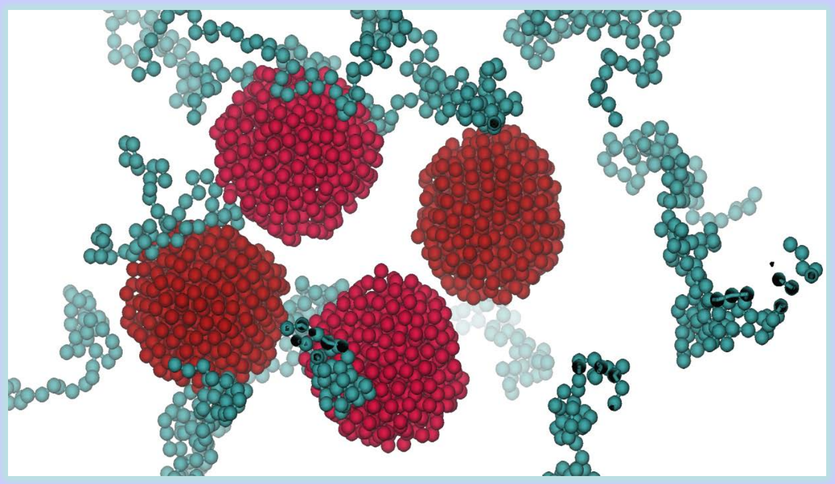
We look at the polymer glass transition in bulk samples, but are especially interested in thin polymer films. At the nanolevel, these films are often comparable to having one molecule-thickness..

In a thin, free-standing film without a substrate, the glass transition temperature is very low, as compared to that of bulk sample, with a difference of fifty-sixty degrees Celsius. It's a huge effect, and we try to understand why, what is so specific about it.



Polymer Nanocomposites

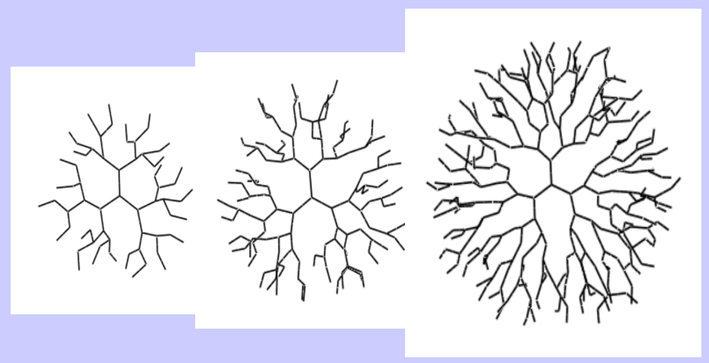
Usually, in these materials inorganic nanoparticles (as Carbon Black or silica, for example), commonly referred to as fillers, are added to polymer matrix in order to increase its rigidity at low strain magnitudes. Nonlinear phenomena restrain the performance of the resulting composites and the development of possible new applications. Our goal is to connect the viscoelastic properties of the polymer composites with their microstructure, and the microstructure with the adhesion polymer-filler interactions at the segmental scale.



Brownian Dynamics: Polymer cauliflower

Brownian dynamics is a simulation technique to study the diffusive behaviour of a system of interacting Brownian particles in solution. In our group we use this approach to perform BD simulations of hyperbranched polymers and their complexes.

A dendrimer (from Greek *dendra* for tree) is an artificially manufactured or synthesized molecule built up from branched units called monomers. In general, these molecules possess a recursively branched, tree-like structure. Hyperbranched molecules are characterized by non-ideally, irregularly branching arrays, in which not every repeat unit contains a branching juncture, resulting in polydisperse shapes. We perform BD simulations of both statistical and dynamical properties of these polymers.



The structures of three hyperbranched molecules with different degree of branching and number of generations

Join us!

Our students benefit from the collaboration with other polymer research groups even far away, e.g. at San Diego University in USA and Moscow State University in Russia. We offer many possibilities for internal, external, and MSc projects:

-Computer simulation of glass transition in amorphous polymers;

-Multiscale modelling of morphologies for new organic solar cells ;

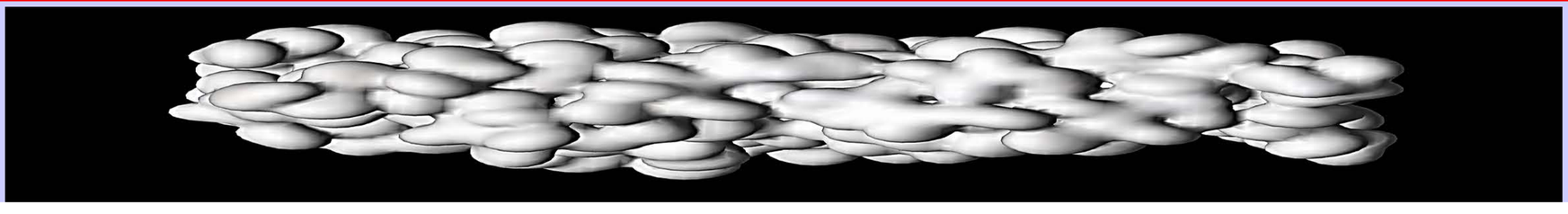
-Multiscale simulations of polyelectrolyte membranes for flow and fuel cells;

-Molecular dynamics of thin polymer films;

-Coarse-grained modelling of hyperbranched polymers and their complexes.

Publications

[1] A.V. Lyulin, B. Vorselaars, M.A. Mazo, N.K. Balabaev, M.A.J. Michels, *Europhys. Lett.* 71, 2005, 618.
[2] A.V. Lyulin, M.A.J. Michels, *Phys. Rev. Lett.* 99, 2007, 085504.
[3] J.-L. Barrat, J. Baschnagel, A. V. Lyulin, *Soft Matter* 2010, 6, 3430.



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