Atomistic modeling of hybrid polymers

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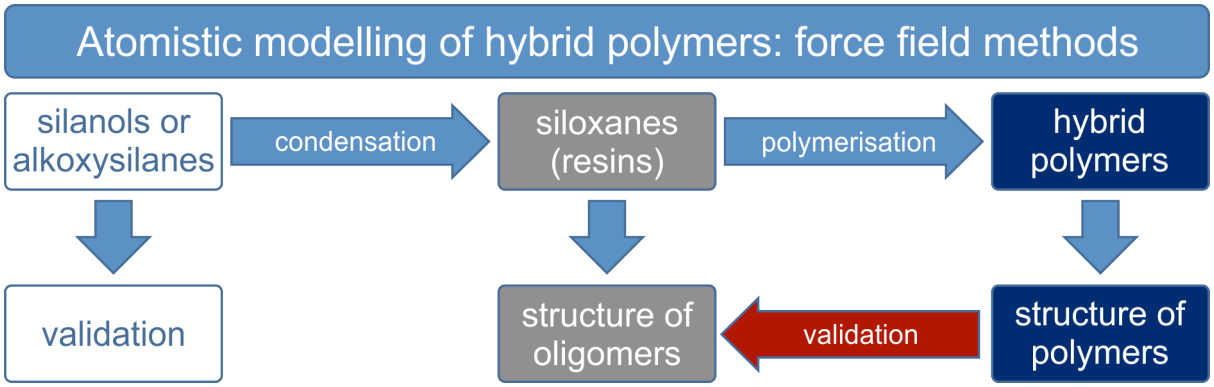
Hybrid polymers are a special class of hybrid materials, not only combining inorganic and organic compounds on a molecular scale, but joining inorganic and organic polymer structures covalently bonded to each other. They are synthesized in a two-step process from silanol and alkoxysilane precursors containing polymerizable functionalities: First, a polycondensation leads to the so-called resin, containing siloxane oligomers. These oligomers are subsequently polymerized, initiated thermally or photochemically, to form the hybrid polymer.

These materials are highly versatile, offering many possible technical or biomedical applications [1]. Except for first basic atomistic modeling studies [2,3], the atomistic structure of the resin or the final hybrid polymer remains often unknown – mainly because the materials are not well defined on a molecular scale and experimental data is very difficult to obtain. Some knowledge on the oligomer structure in the resin can be obtained by ^{29}Si-NMR spectroscopy, while the degree of conversion in the polymerization reaction can be determined by Raman spectroscopy.



The vast amount of possible oligomeric species and their large size limit the simulations on an atomistic scale to force field methods. However, Monte Carlo and Molecular Dynamics methods can be applied for model generation and investigations on material properties at ambient conditions.

Over the last years, we developed strategies and methods to perform force field modeling studies on these complex materials, a general approach is shown in the flow-chart above. The materials investigated differ in the number and type of precursors utilized, but follow the described two-step procedure. Depending on the number of different oligomeric species, we present alternative strategies to handle the process of model generation. They all have in common the large number of models to be considered. The structure of the polymer – and in particular the shrinkage behavior – allows to draw conclusions on the resin models evaluated.

We chose to design our simulations and strategies to be employable at comparably low computational cost, making the strategies presented available to many modeling scientists and even experimental scientists without access to large cluster systems.

[1] C. Sanchez et al., *Chem. Soc. Rev.*, **2011**, *40*, 696–753.
[2] F. Burmeister et al., *Optically Induced Nanostructures*, **2015**, 239–266.
[3] S. Fessel et al., *J. Sol-Gel. Sci. Technol.*, **2012**, *63*, 356–365.