Towards the coarse-grained modelling of dimethacrylate-based biomaterials

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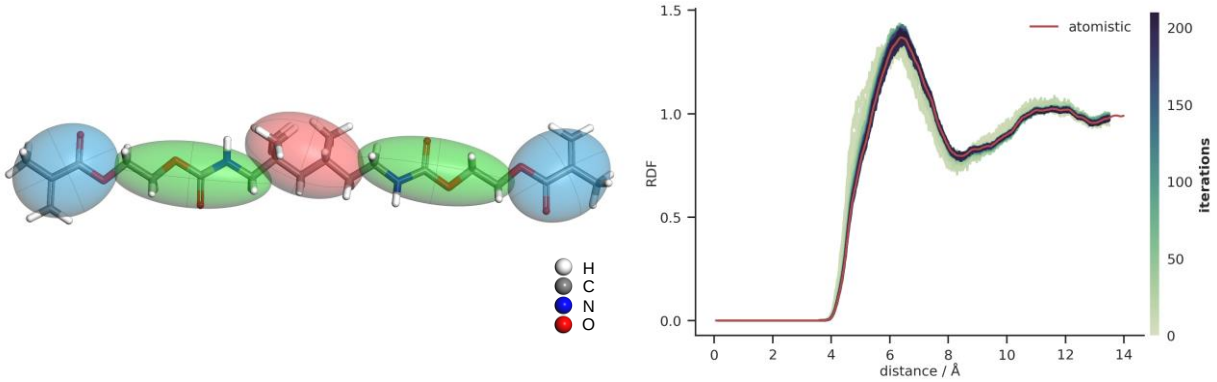
In restorative dentistry dimethacrylate-based composites are a prominent class of biomaterials due to their clinical processability and aesthetic properties. For this purpose, the monomer resins can be cured via photo-induced radical polymerisation. However, a typical decrease in volume with increasing conversion of double bonds during polymerisation is a major drawback affecting longevity of implants. The so-called polymerisation shrinkage can be compensated by both, tuning of steric hindrance of the polymer chains as well as incorporation of filler materials. [1]



For polymers, computer based simulation methods offer powerful tools for the investigation of structural and mechanical properties. In the case of polymers, the simulation requires sufficiently large ensembles of atoms leading to long calculation times on the atomistic scale. A possible solution to this challenge is the application of so-called coarse-grained (CG) models, which provide acceleration by reducing the degrees of freedom in the system. This is realised by the replacement of whole molecules or fragments by single interaction sites, often called beads or superatoms.

The parameters for coarse-grained models can be derived from atomistic reference models or experimental data. As a model system, urethane dimethacrylate (UDMA) was chosen, which is already used in dental applications nowadays. The model consists of three different types of beads for the monomer, in the left figure indicated by ellipsoids. The terminal methacrylate groups are the reactive species regarding polymerisation and are therefore defined as an individual bead type. The remaining types are chosen in a way to offer a nearly similar masses. For oligo- and polymers an additional type for the backbone is introduced. The approach followed in our work is the iterative Boltzmann inversion, where the parameters are optimised against local structures from atomistic molecular dynamics simulations e.g. radial distribution functions (RDF). [2] The sequential convergence of an example RDF is shown in the figure (right).

Employing the generated parameters, we aim for investigation of the polymer structure on the mesoscale with focus on cavity formation during polymerisation, which contribute to the reduction of shrinkage. In the future, we intent to study dimethacrylate-polymers with various composition, utilizing the combining rules to derive parameters from neat systems. [3]

[1] R. Sakaguchi, J. Powers, *Craig's restorative dental materials*, Elsevier/Mosby, 2012.
[2] V. Rühle, C. Junghans, A. Lukyanov, K. Kremer, D. Andrienko, *J. Chem. Theory Comput.*, **2009**, *5*, 3211-3223.
[3] H.J. Qian, P. Carbone, X. Chen, H. A. Karimi-Varzaneh, C. C. Liew, F. Müller-Plathe, *Macromol.*, **2008**, *41*, 9919-9929.