









Understanding the effect of coarse grained mapping on the properties of amorphous polyethylene terephthalate

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INTRODUCTION & MOTIVATION

To date, the food industry is predominated by the use of flexible multiwall packages due to their excellent mechanical and thermophysical properties. Nonetheless, this is followed by a poor recyclability rate related to the increased cost in material separation, which results into a considerable plastic pollution to the environment.

Computer simulations offer an excellent platform to design novel materials and provide guidance to experiment. However, despite recent advances, it is still very challenging to establish direct relationships between local chemical details and the macroscopic properties. The reason lies in the fact that the polymer dynamics spans over a wide range of time scales, ranging from femtoseconds for local chain vibrations to seconds for chain relaxation and diffusion.¹

In this regard, coarse grained (CG) models have drawn considerable attention to bridge the differences in time scales.² CG models neglect the fastest degrees of freedom by grouping together multiple atoms into a single CG interaction site (bead), flattening the rugged potential energy surface (PES). This allows the use of larger time steps for integrating the equations of motion, thereby achieving longer simulation times.

Our goal is to develop an automatized multiscale workflow to study amorphous polyethylene terephthalate (PET) from full atom to CG molecular dynamics. Ultimately, we aim to extend it to other polymers such as PEF, PLA and cellulose, as well as to the crystalline phase.

METHODOLOGY

Full atom molecular dynamics

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Quantum mechanically parametrized force field (AA-QMD)³ vs OPLS/AA (Ligpargen)⁴

Melt-quench-annealing protocol to obtain amorphous state⁵

Structural data collected with 20 independent production runs of 100 ns in NPT ensemble at 1 atm and 298 K

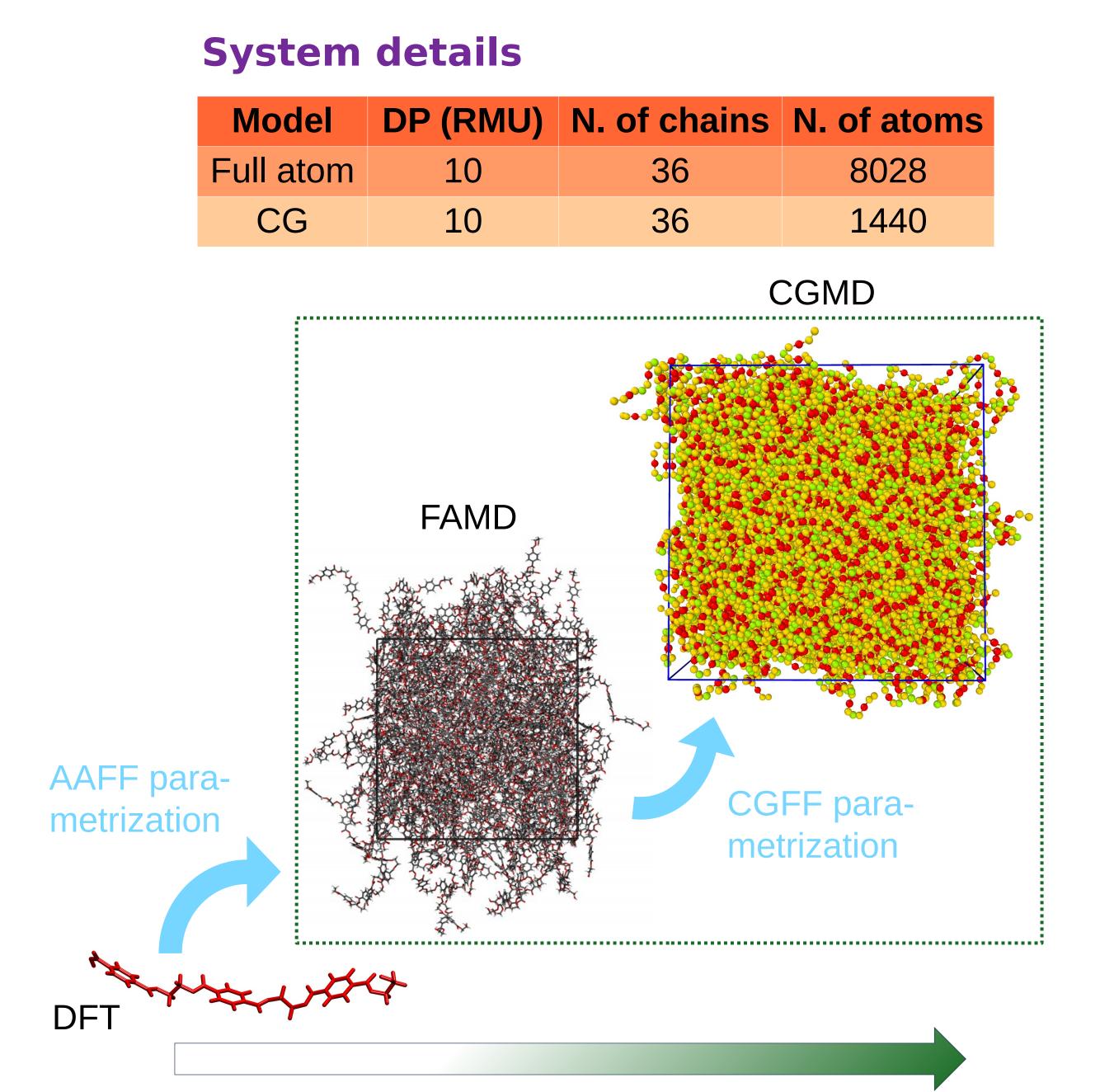
Leap-Frog algorithm to integrate EOMs with a time step of 1 fs

Coarse grained molecular dynamics

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Structural data collected with 10 independent production runs of 50 ns in Langevin dynamics at 298 K

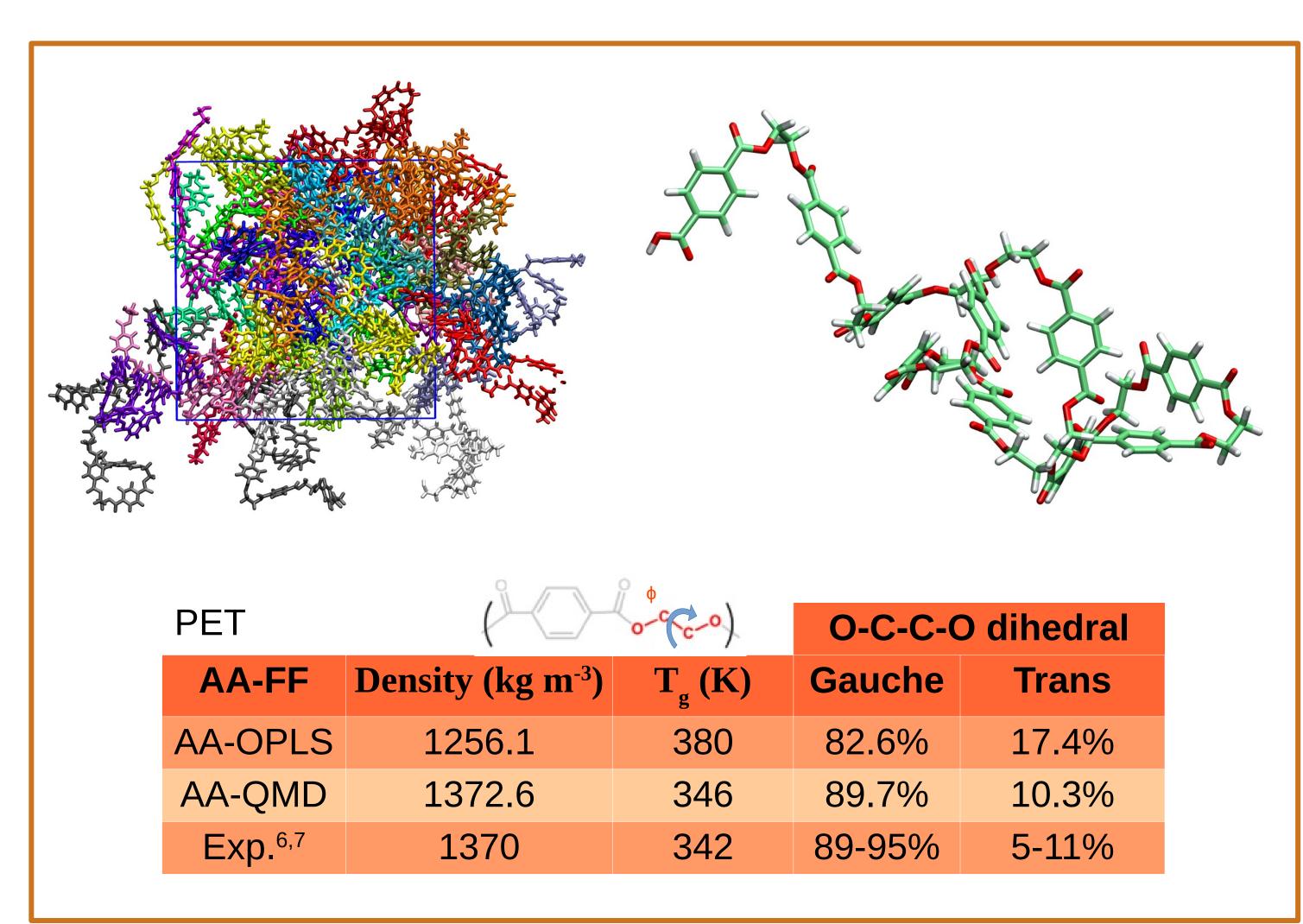
Verlet algorithm to integrate EOMs with a time step of 5 fs



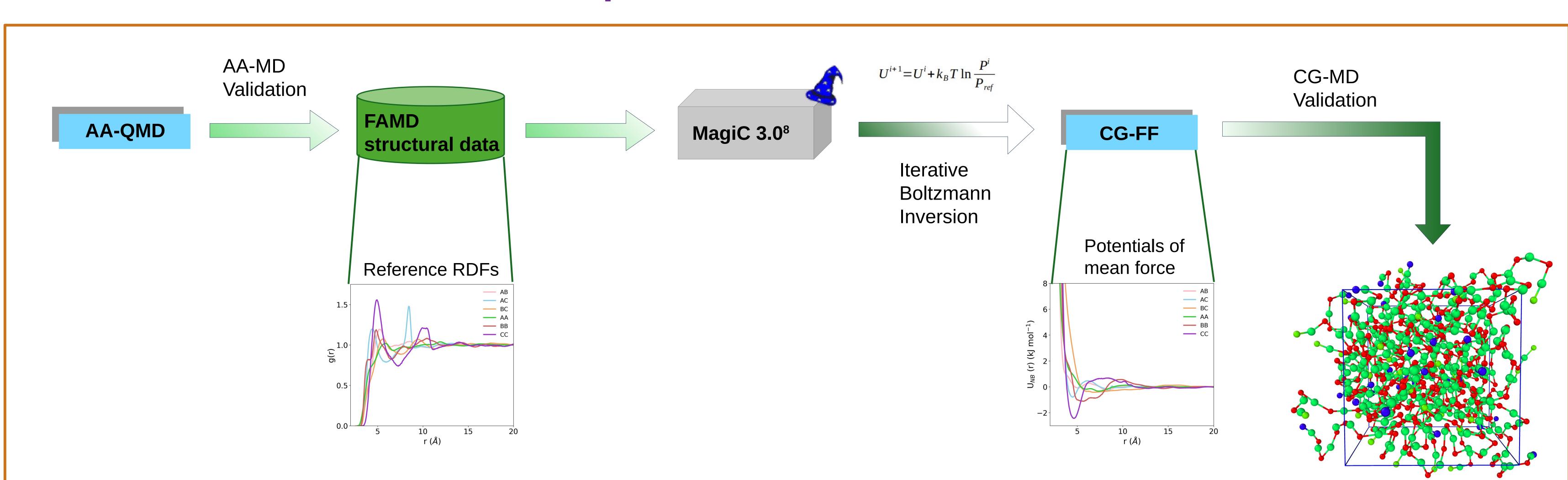
Length scale

RESULTS & DISCUSSION

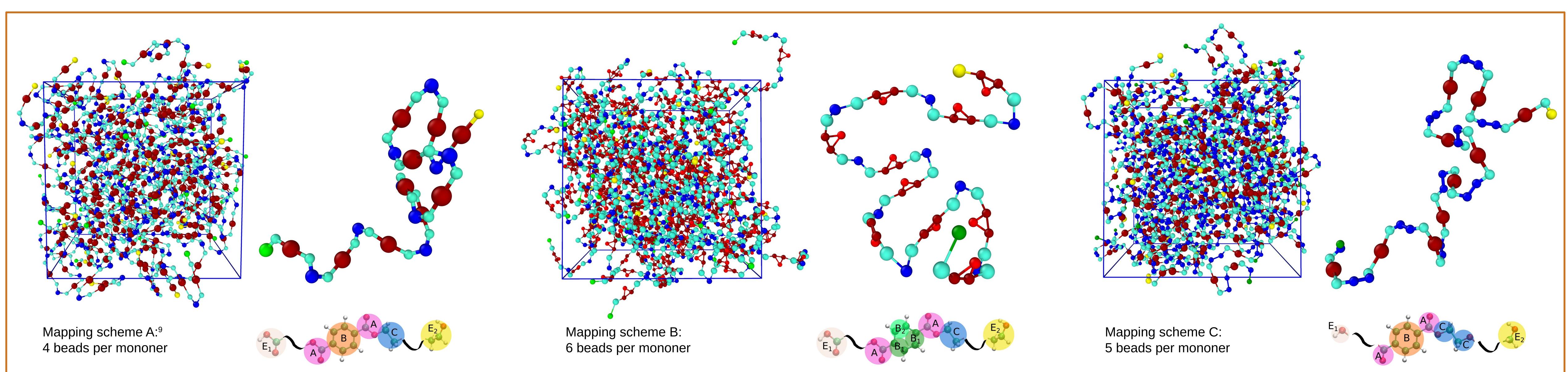
Full Atom Molecular Dynamics (FAMD)

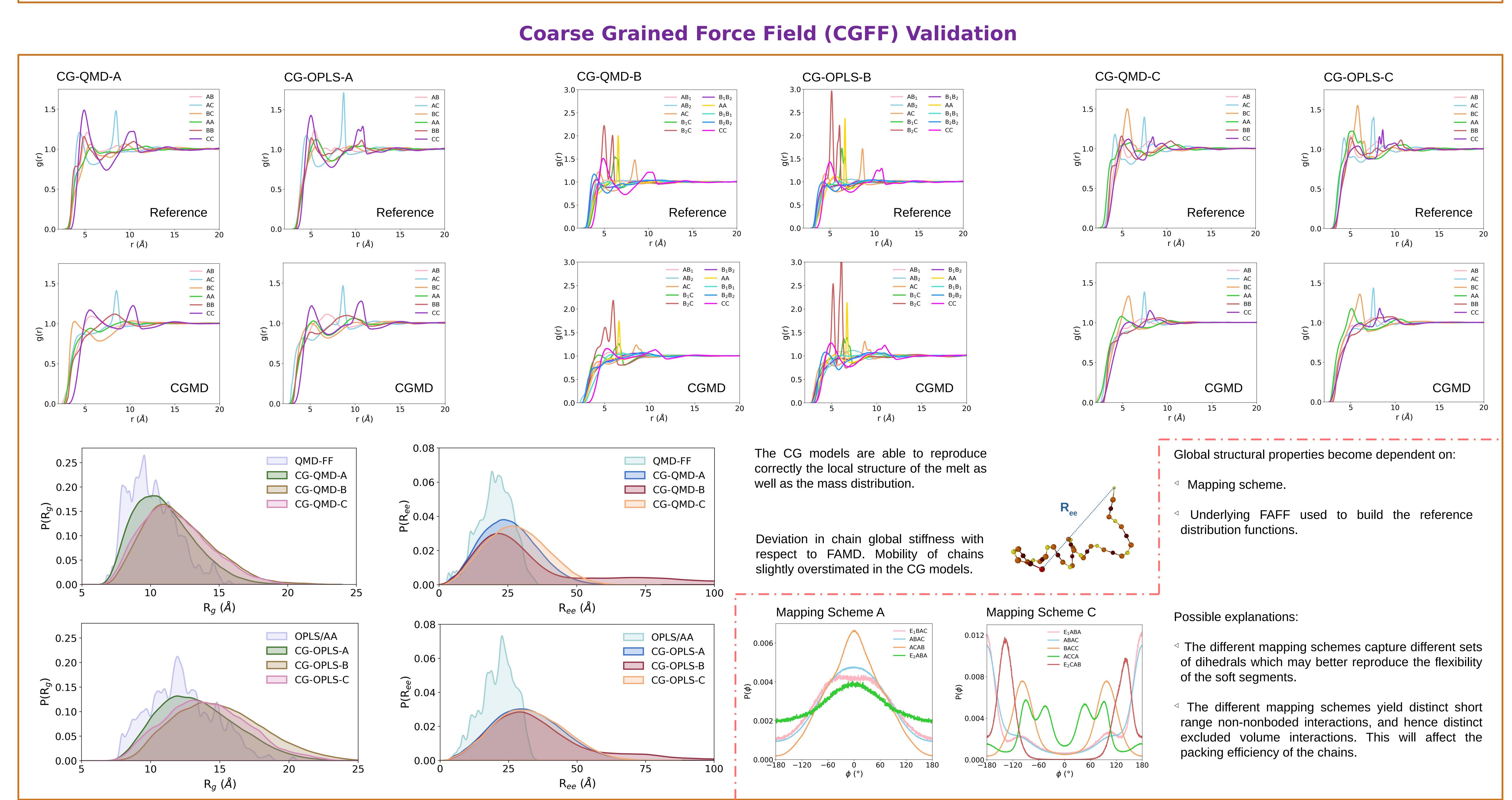


Implementation & Workflow



Coarse Grained Models





CONCLUSIONS

- In this work we have devised a **multiscale workflow** to study amorphous polymers from full atomistic to coarse grained models.
- spatiotemporal scales and predict physicochemical properties.

Our results show that multiscale modeling is a powerful technique to bypass motions of large

FAMD: QMD-FF improves density and global chain stiffness with respect to AA-OPLS.

excluded volume effects.

- **CGMD:** The CG-FF developed with IBI correctly reproduces local structural properties, but introduces some deviations in the global chain stiffness.
- For future work, we will try other parametrizations techniques such as force matching. In addition, we will describe the CG beads with **Gay-Berne ellipsoids** instead of isotropic spheres to address better the

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