

Molecular Simulation Study of the Volume Transition of Hydrogels

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Hydrogels

What is a Hydrogel?

- Three dimensional hydrophilic polymer network
- Most important property: Swelling in aqueous solutions



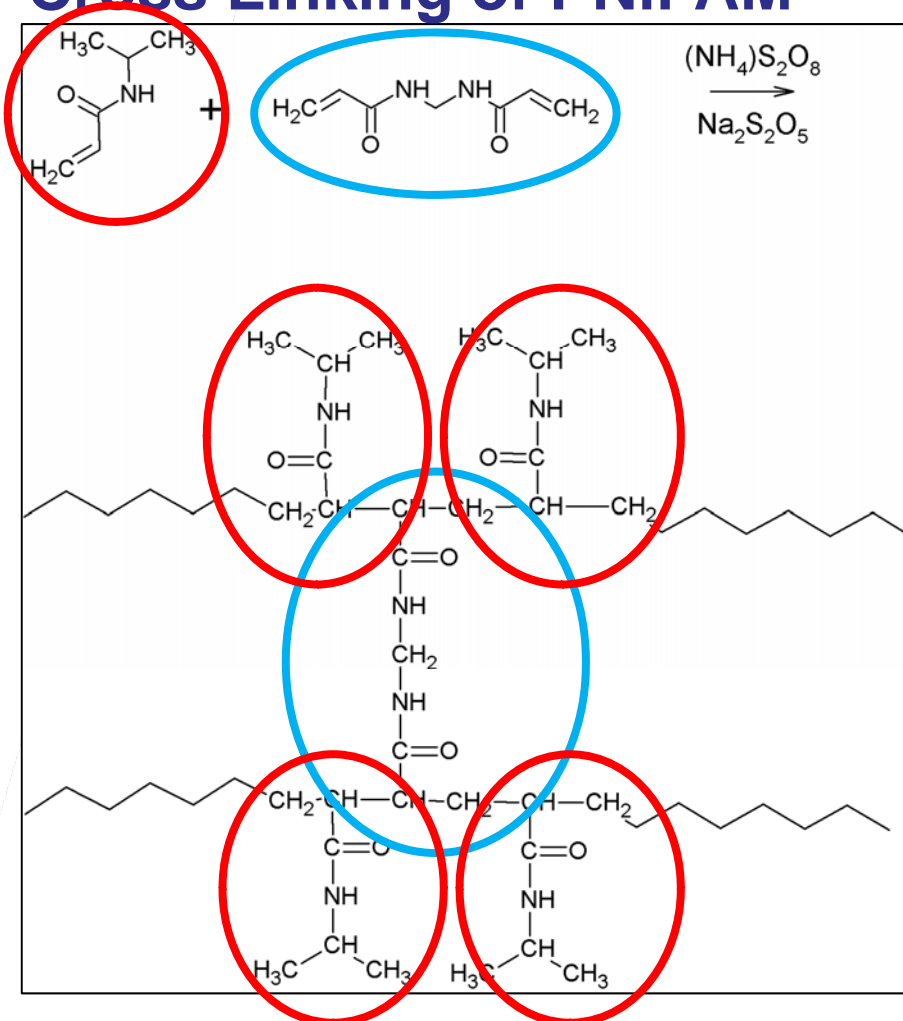
Examples for Applications:

- Super absorber
- Contact lenses
- Drug delivery systems
- Sensors
- Actors (e.g. micro valves)
- Biocatalysis



Poly(N-isopropylacrylamide) (PNiPAM)

Cross Linking of PNiPAM



Single Chain

- Hydrophilic monomers

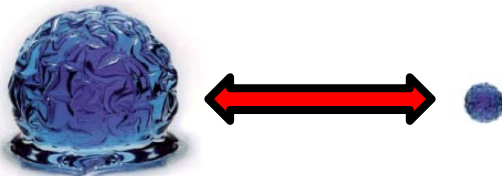
Cross Linker

- Chemical
- MBA

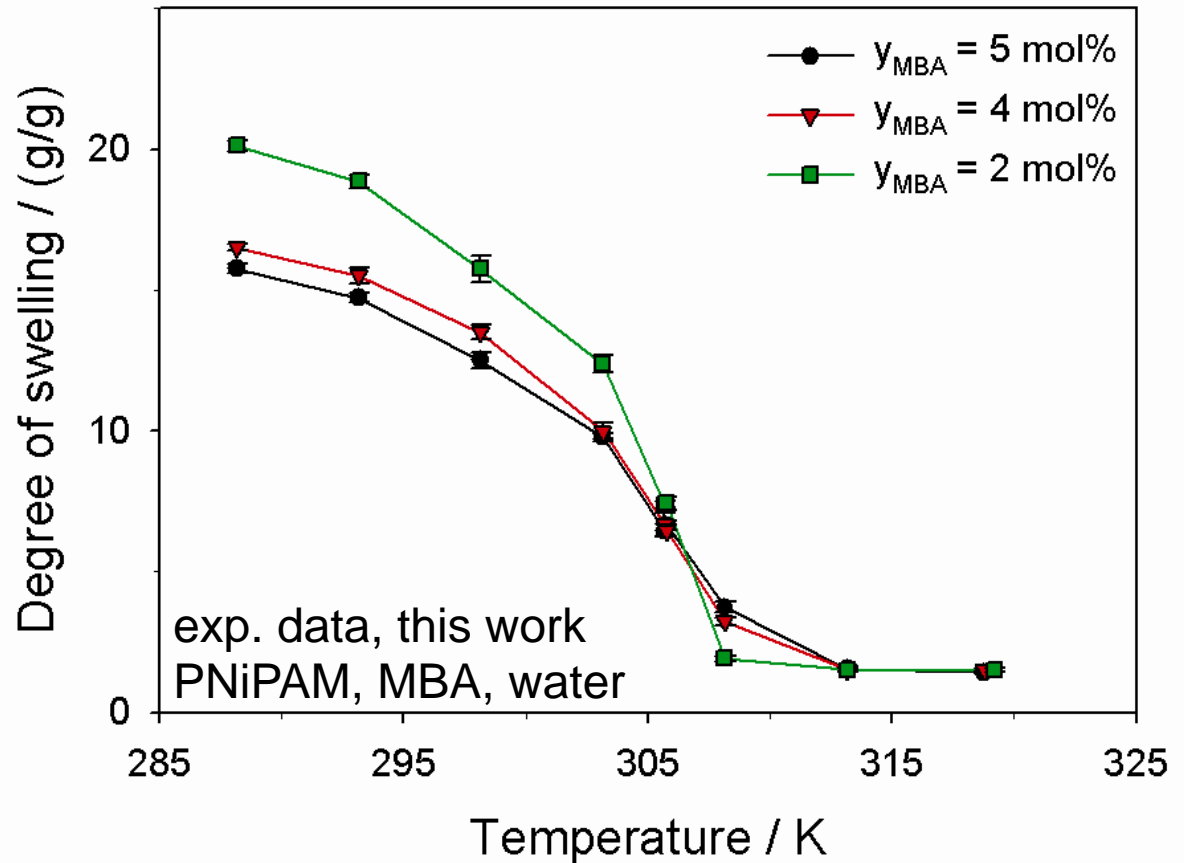
Degree of swelling in aqueous solutions

Degree of Swelling depends on:

- Monomer
- Co-monomer(s)
- Cross linker amount
- Solvent
- pH-value
- Salt concentration
- Temperature
- ...



Influence of the Temperature



Molecular Modeling and Simulation

Modeling:

- Force fields from literature

PNiPAM	water: SPC/E	water: TIP4P
Gromos87 UA	✓	✓
Gromos96 53a6 UA	✓	✓
OPLS AA	✓	✓

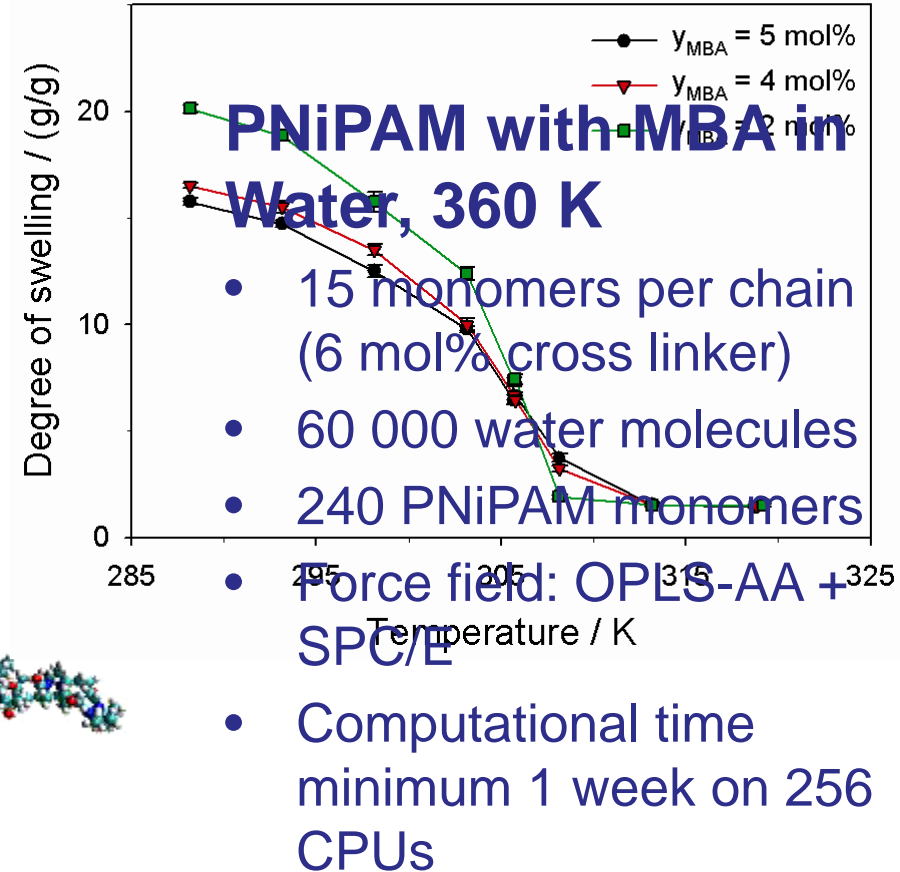
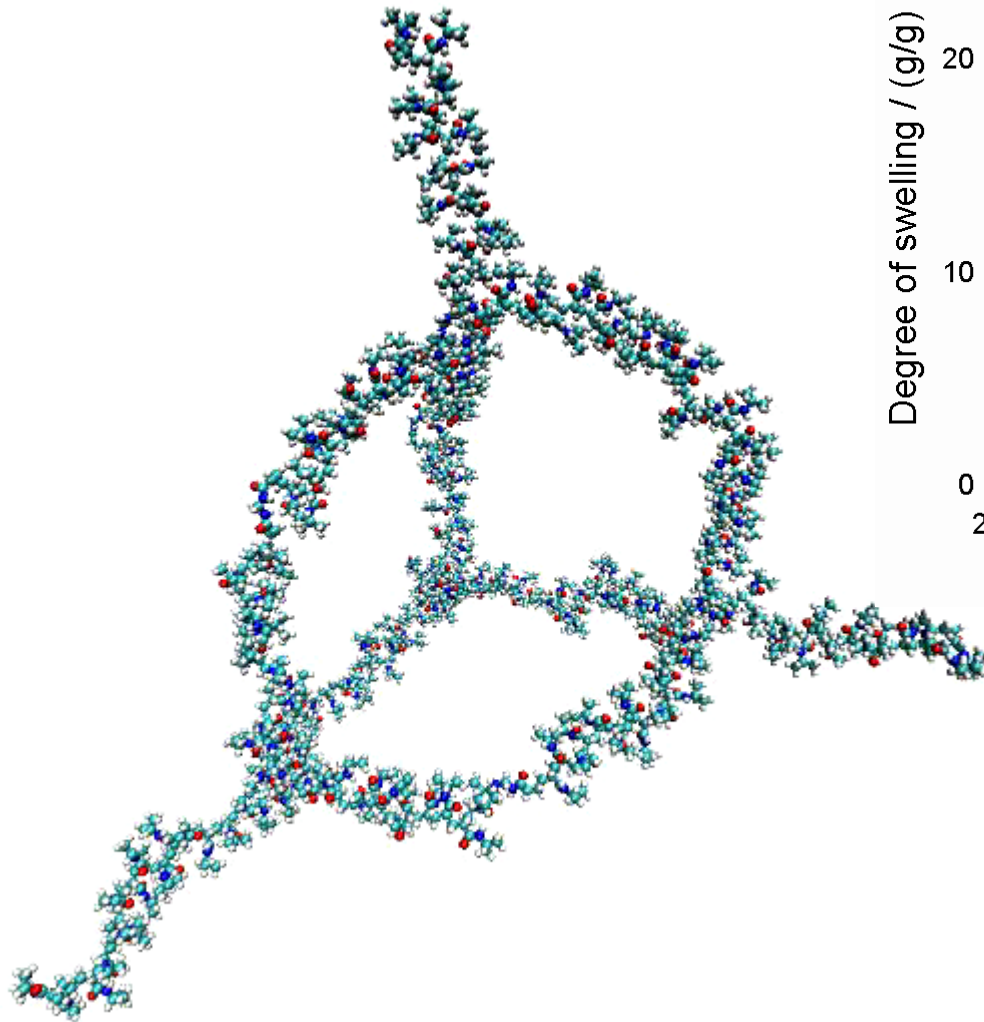
Simulation:

- Molecular dynamics (MD)
- NpT-ensemble

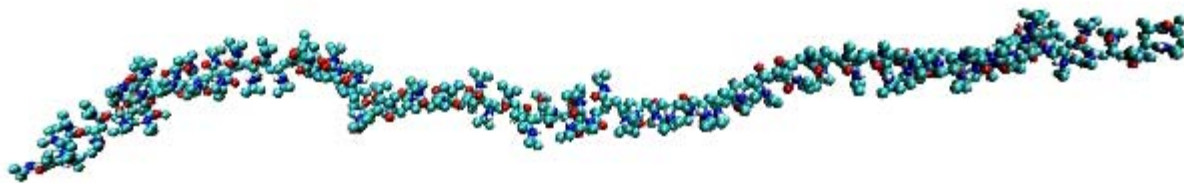
Program:

- Gromacs 4.0.x

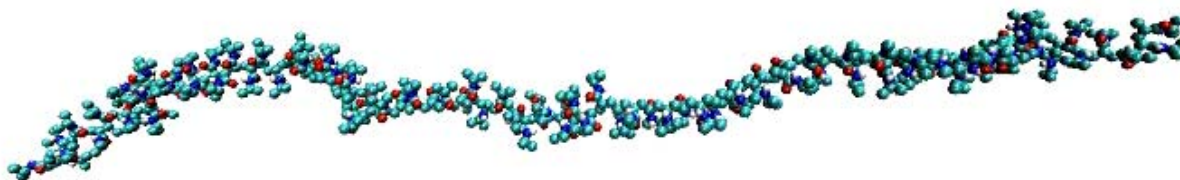
MD-Simulation Hydrogel Network



MD-Simulation Single Chain



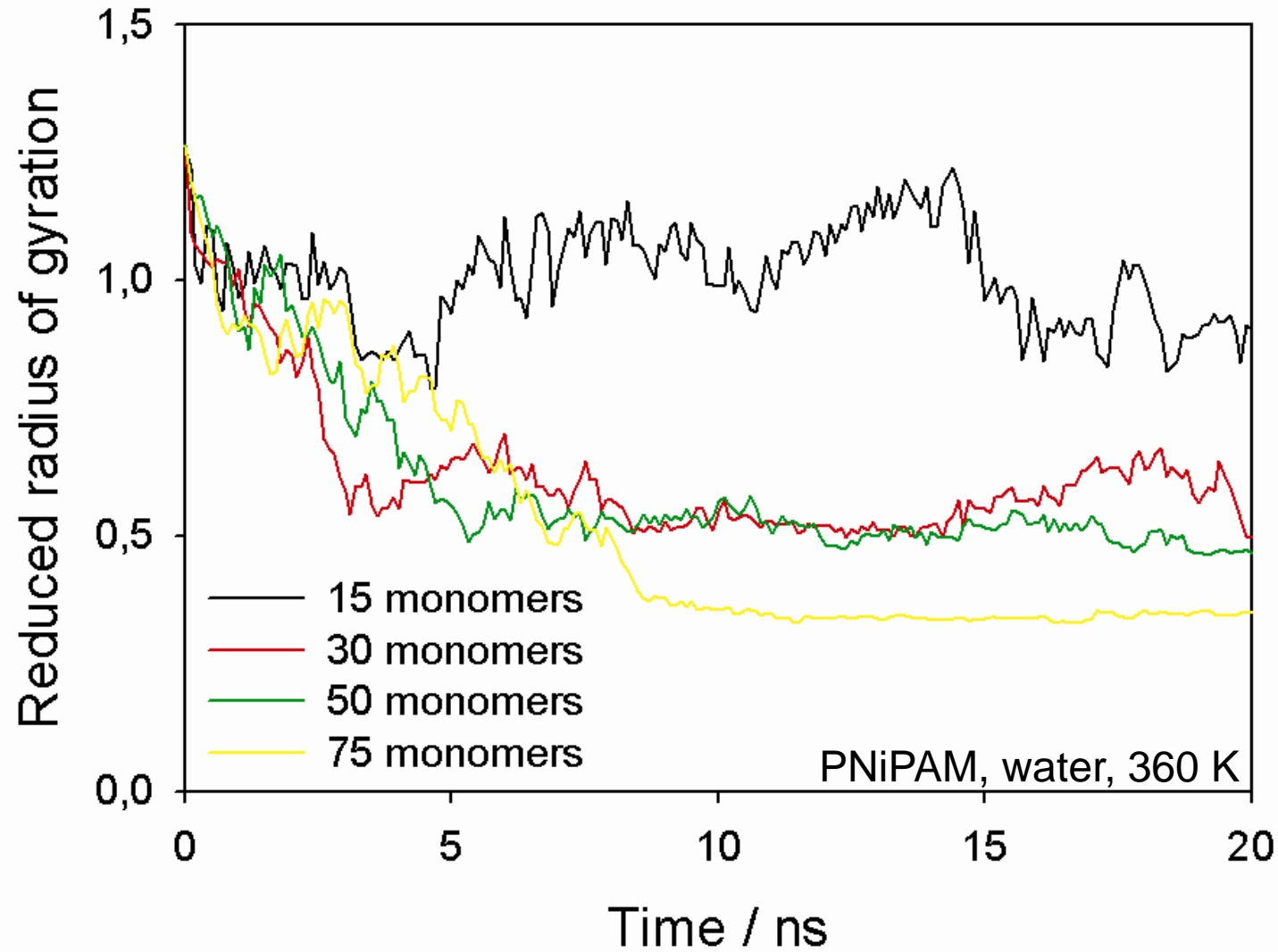
280 K



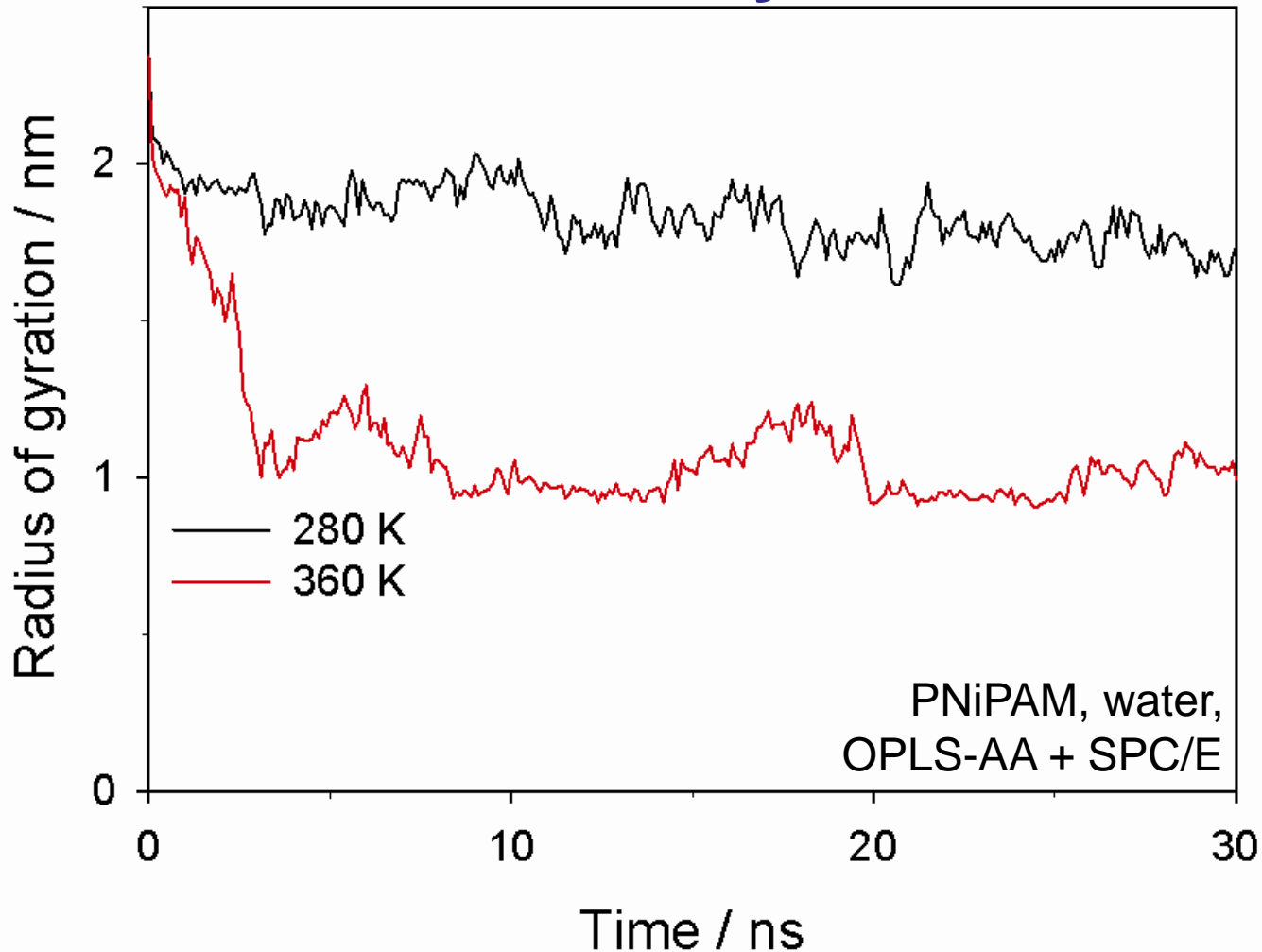
360 K

PNiPAM, water, OPLS-AA + SPC/E, 75 monomers

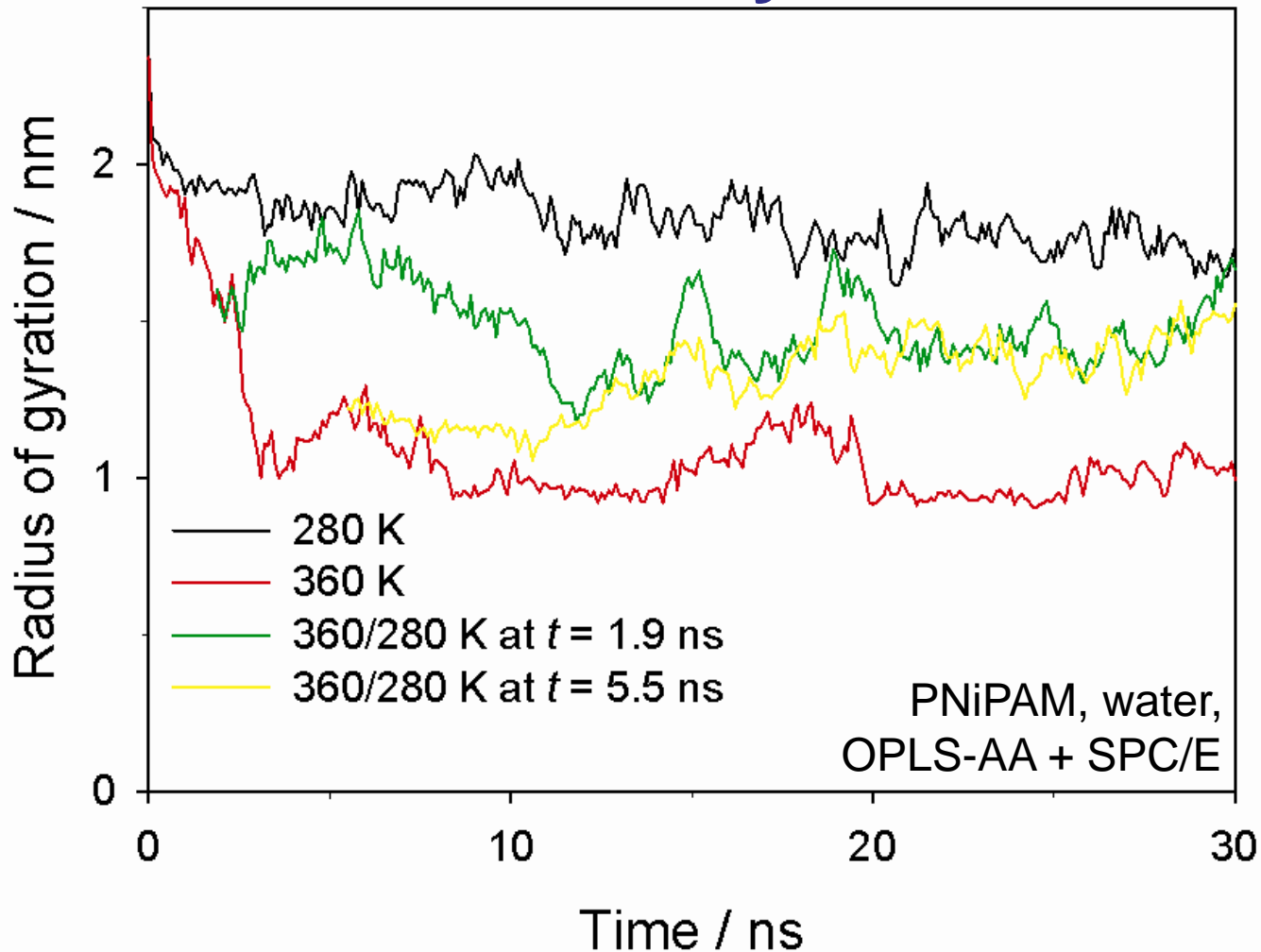
Preliminary Study: Chain Length



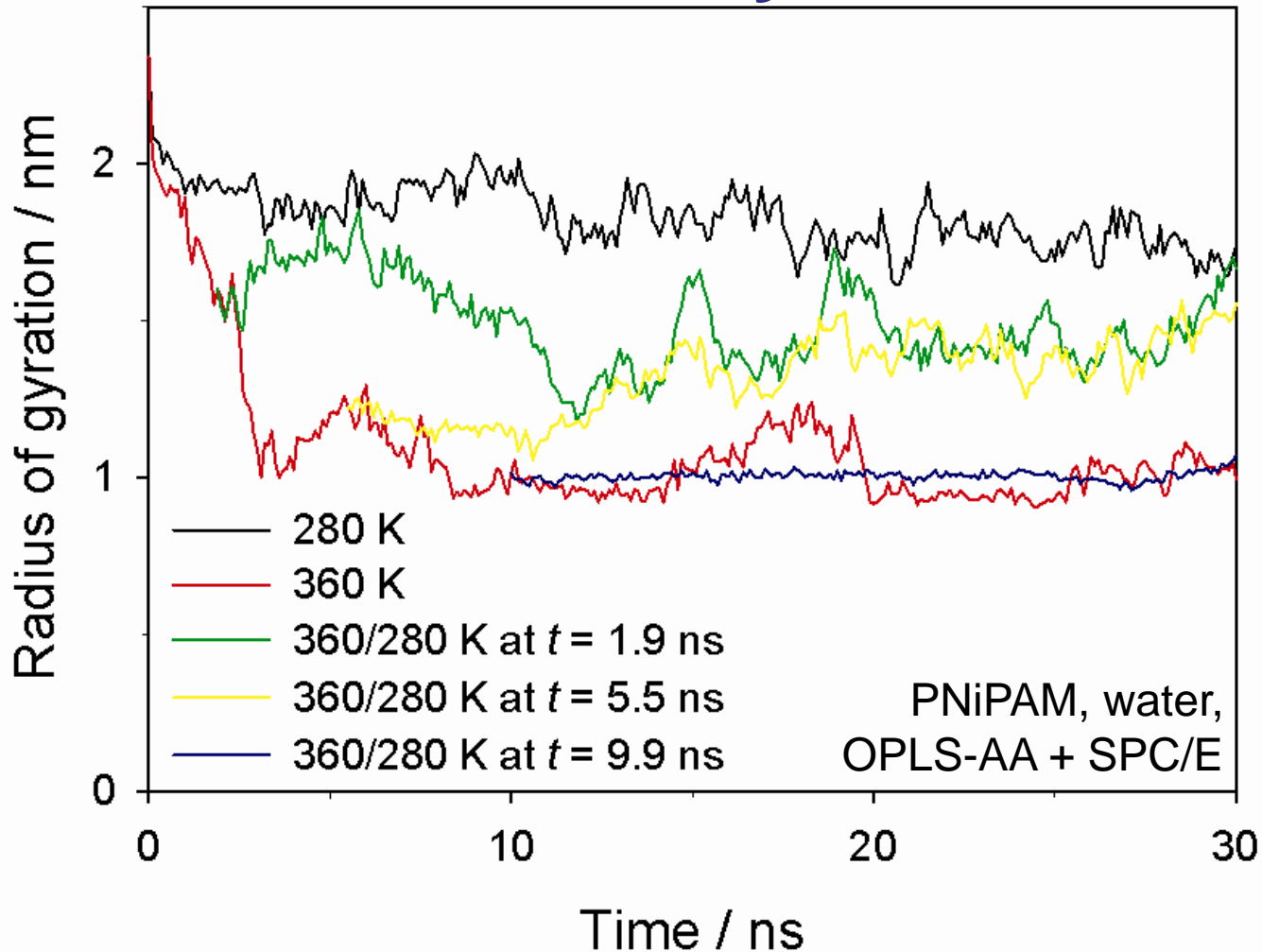
Temperature Dependence of Single Chain Radius of Gyration



Temperature Dependence of Single Chain Radius of Gyration



Temperature Dependence of Single Chain Radius of Gyration





Force Field Comparison - Results

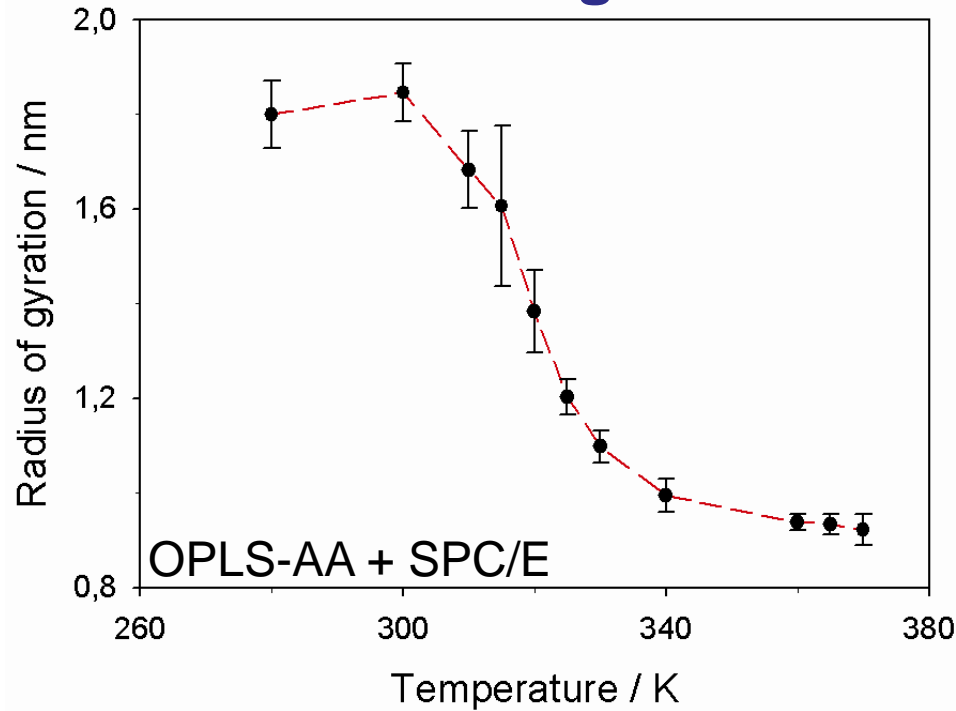
PNiPAM	water: SPC/E	water: TIP4P
Gromos87 UA	-	-
Gromos96 53a6 UA	-	+
OPLS AA	+	+

Legend

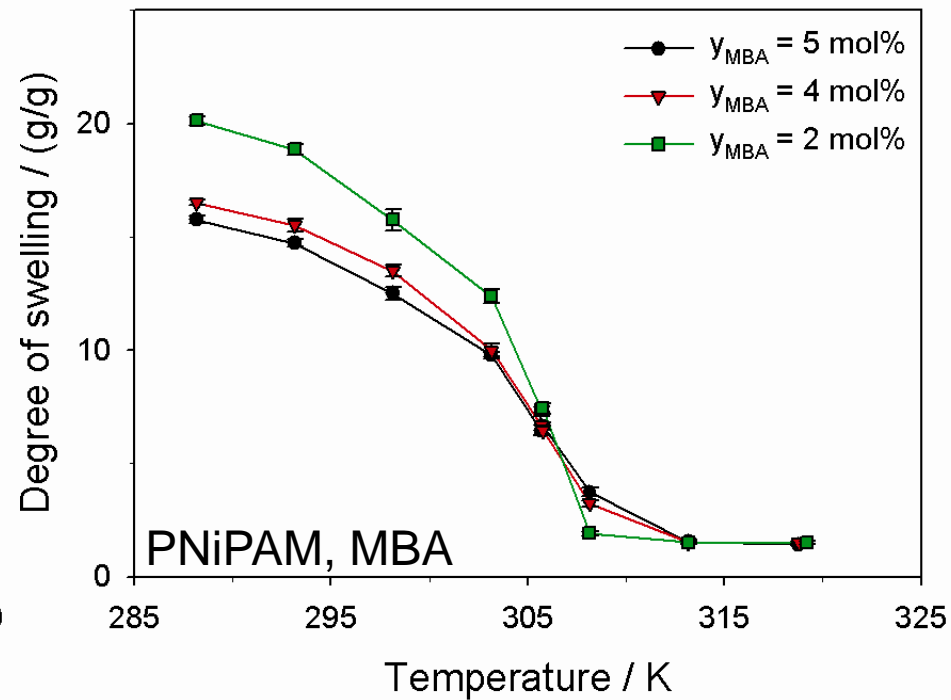
- Force field does not predict effect
- + Force field predicts effect

Temperature Dependence of the Swelling of PNiPAM in Water

Simulation Single Chain



Experiment Hydrogel



Summary

- Swelling of hydrogels: PNiPAM
- Molecular modeling and simulation
- Temperature dependence of swelling
- Hydrogel swelling simulations:
 - Networks
 - Single chains
- Qualitative predictions with force fields from the literature
- Development of force fields for quantitative predictions



Acknowledgements:



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