



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:

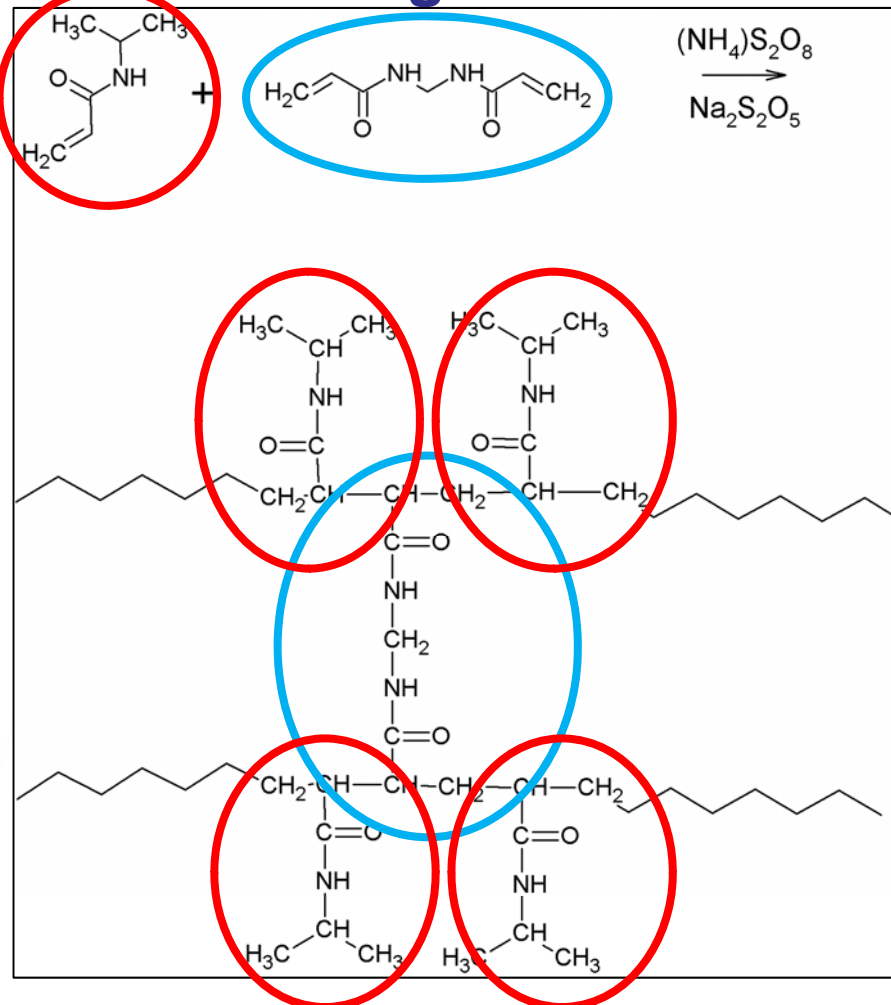
- Super absorber
- Contact lenses
- Drug delivery systems
- Sensors
- Actuators (e.g. micro valves)
- Biocatalysis





Poly(N-isopropylacrylamide) (PNiPAM)

Cross Linking of PNiPAM



Single Chain

- Hydrophilic monomers

Cross Linker

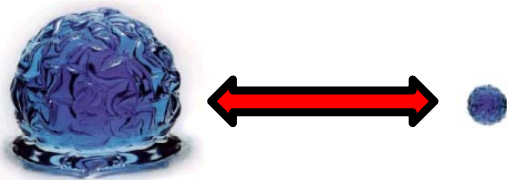
- Chemical
- MBA



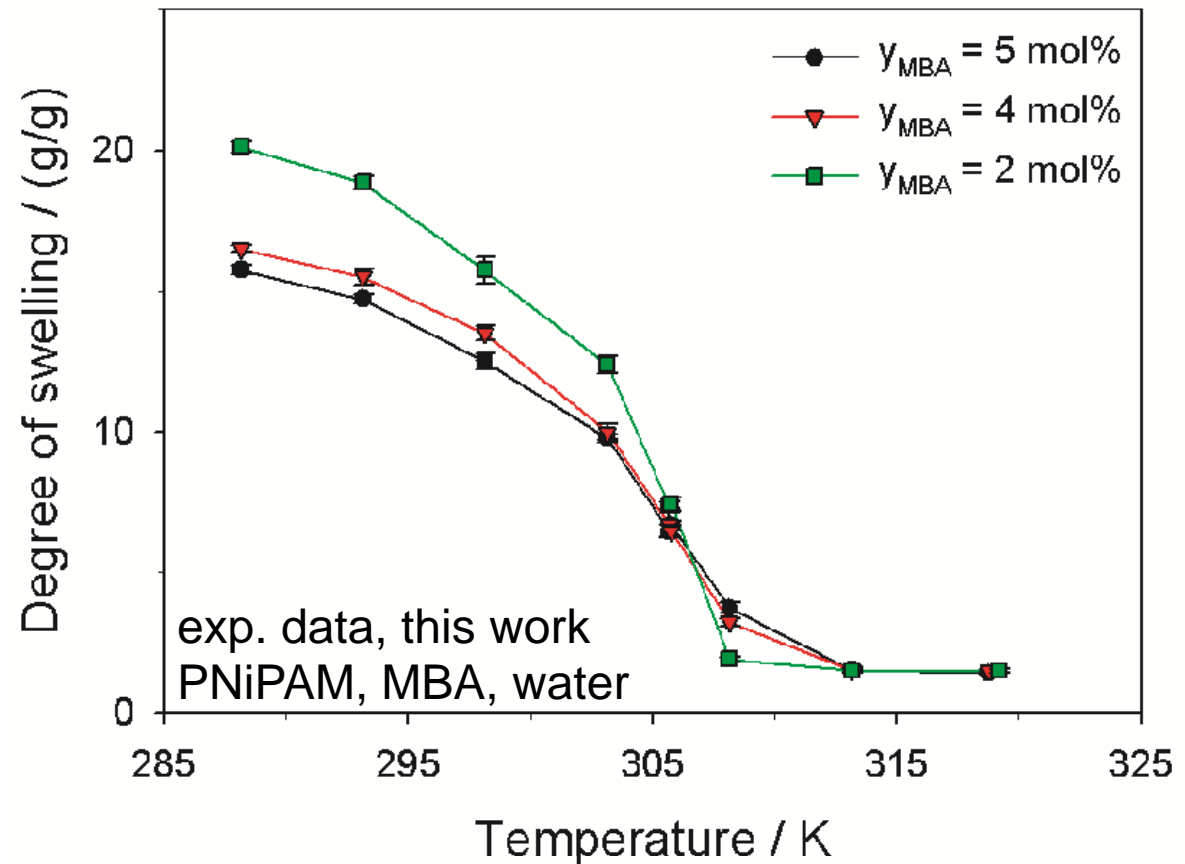
Degree of swelling in aqueous solutions

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:

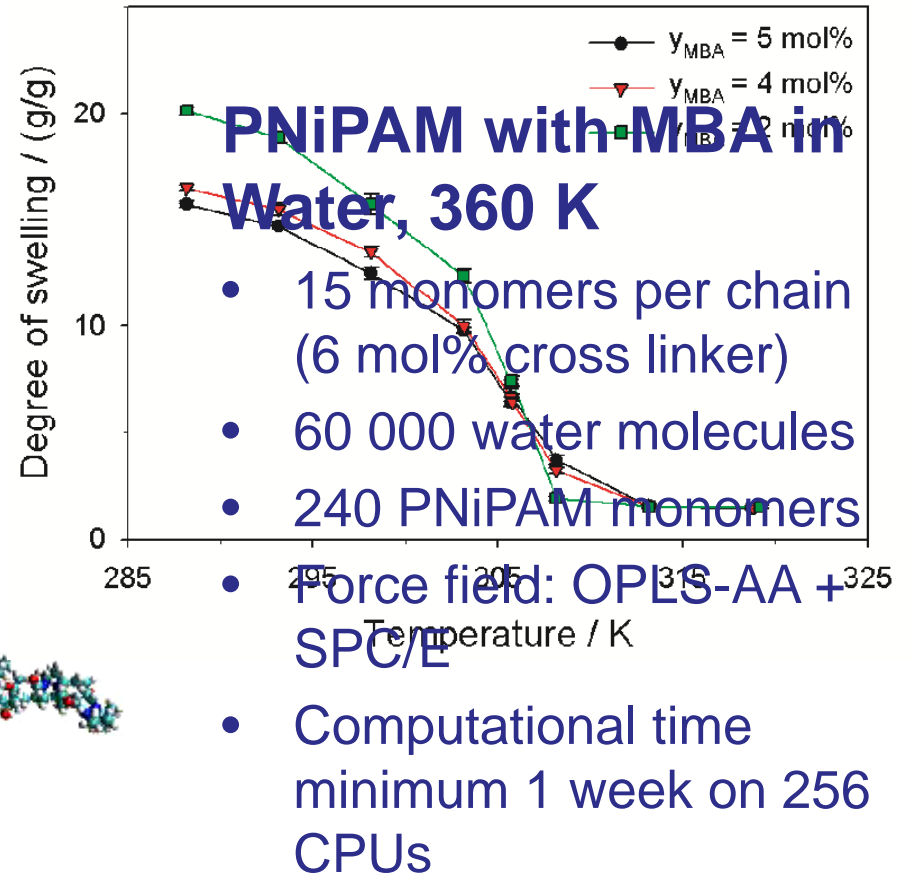
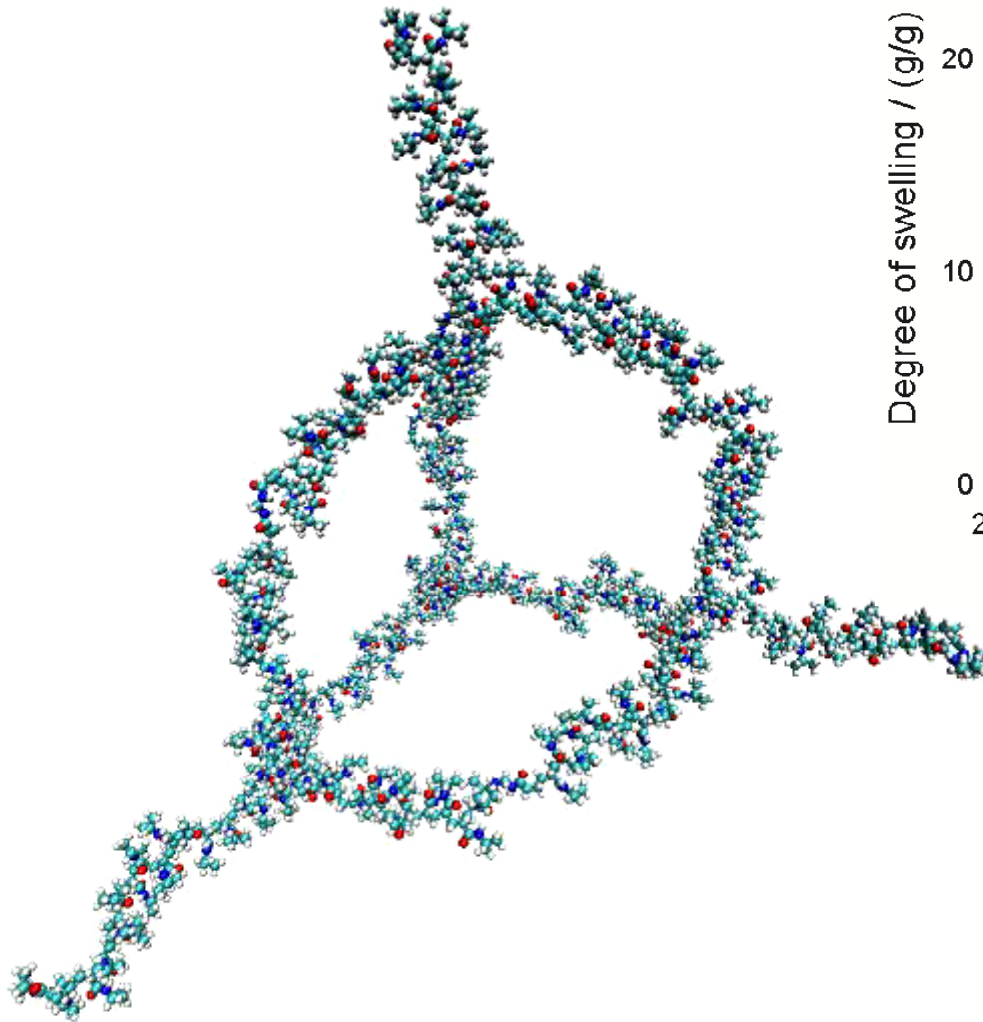
- Molecular dynamics (MD)
- NpT-ensemble

Program:

- Gromacs 4.0.x

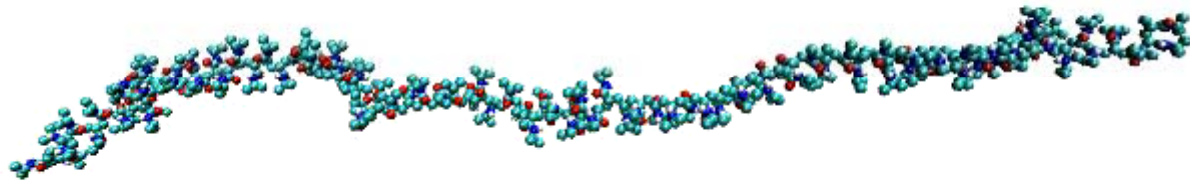


MD-Simulation Hydrogel Network

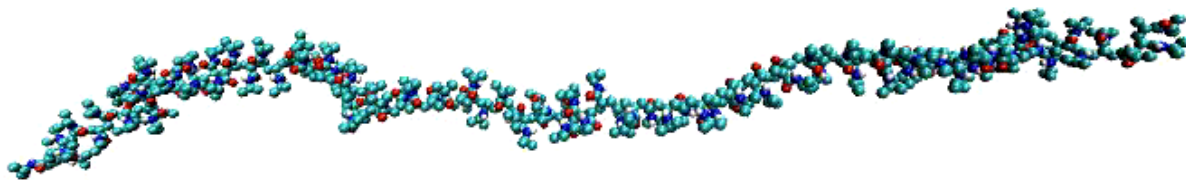




MD-Simulation Single Chain



280 K

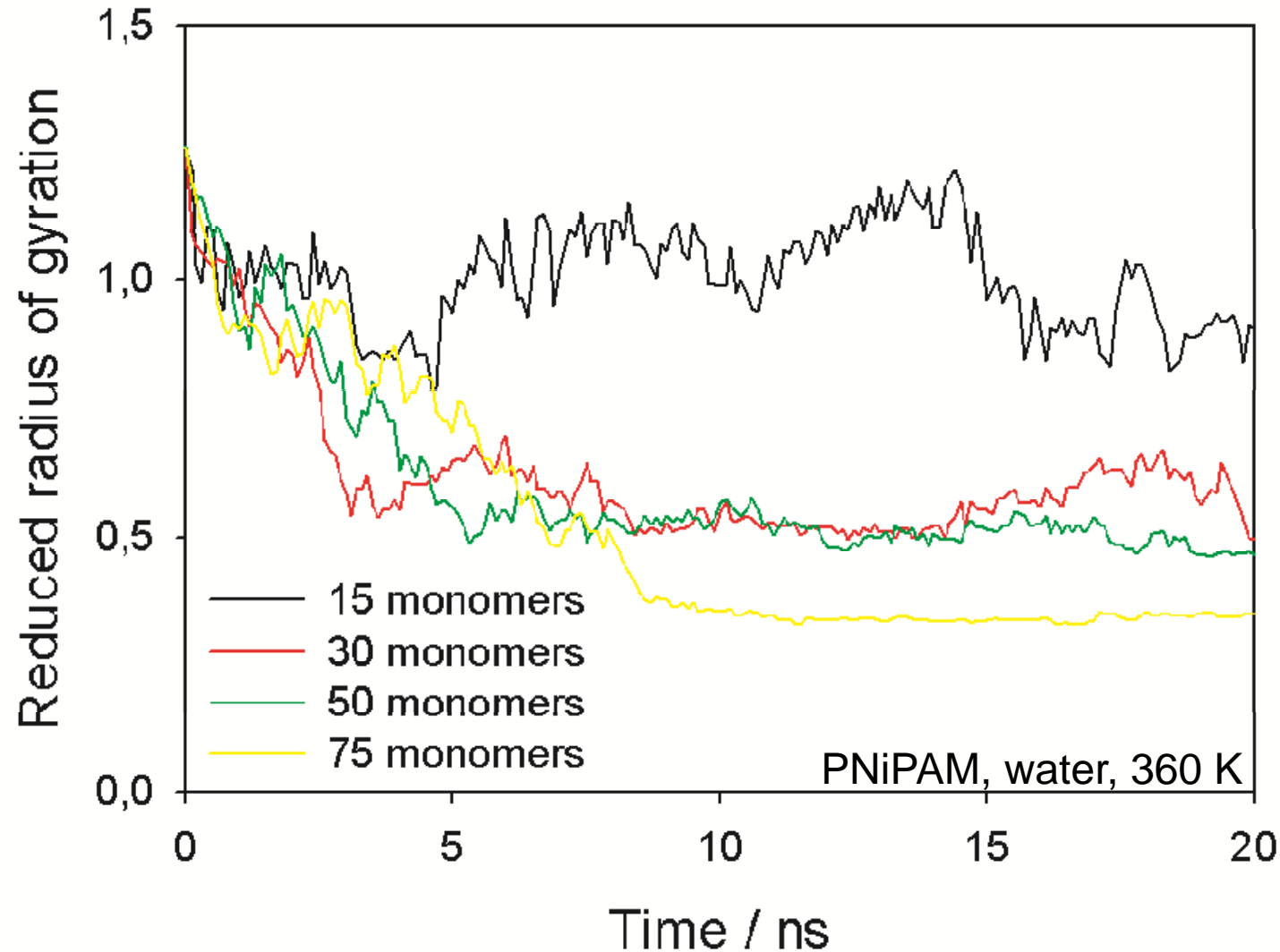


360 K

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

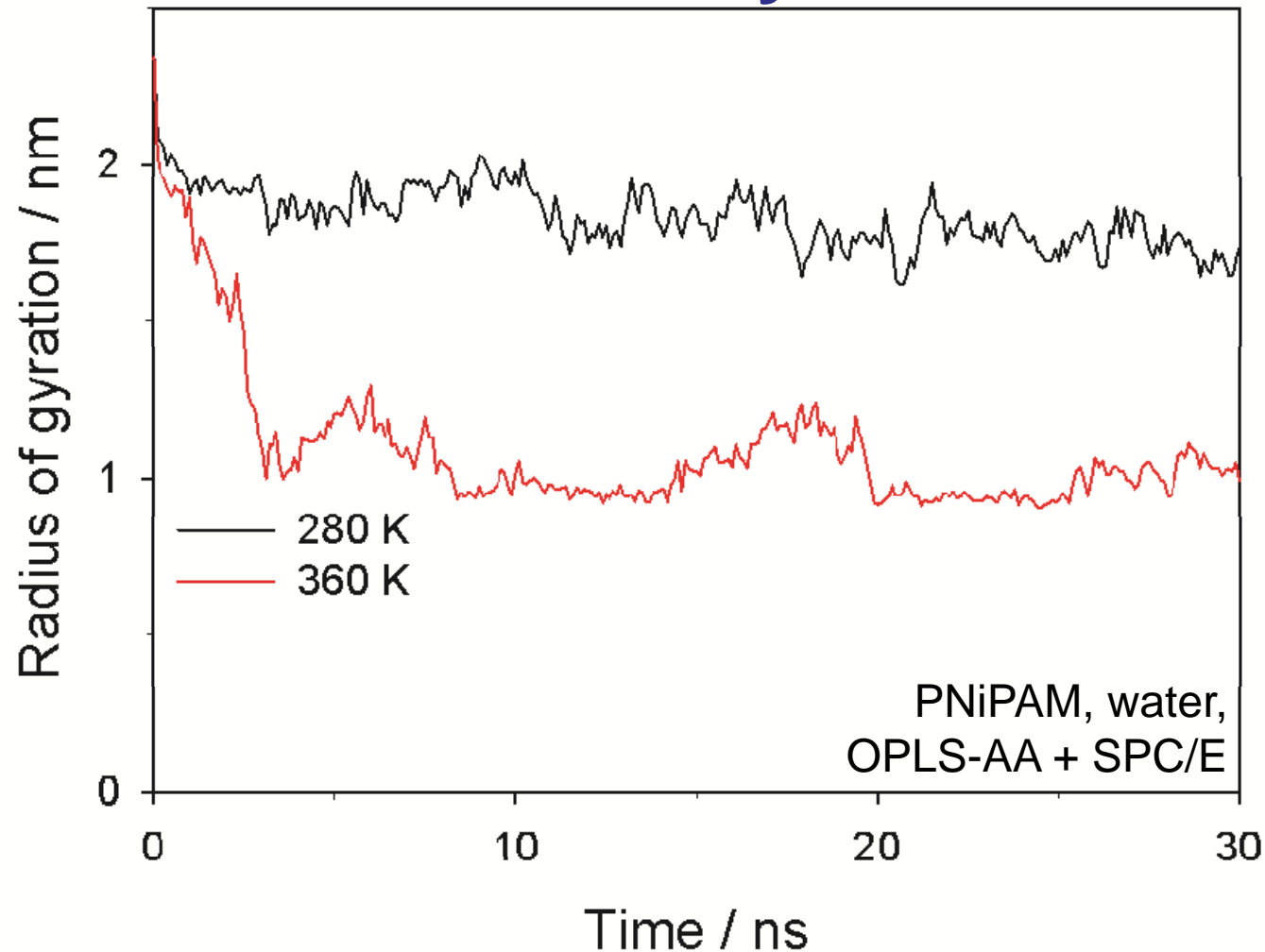


Preliminary Study: Chain Length



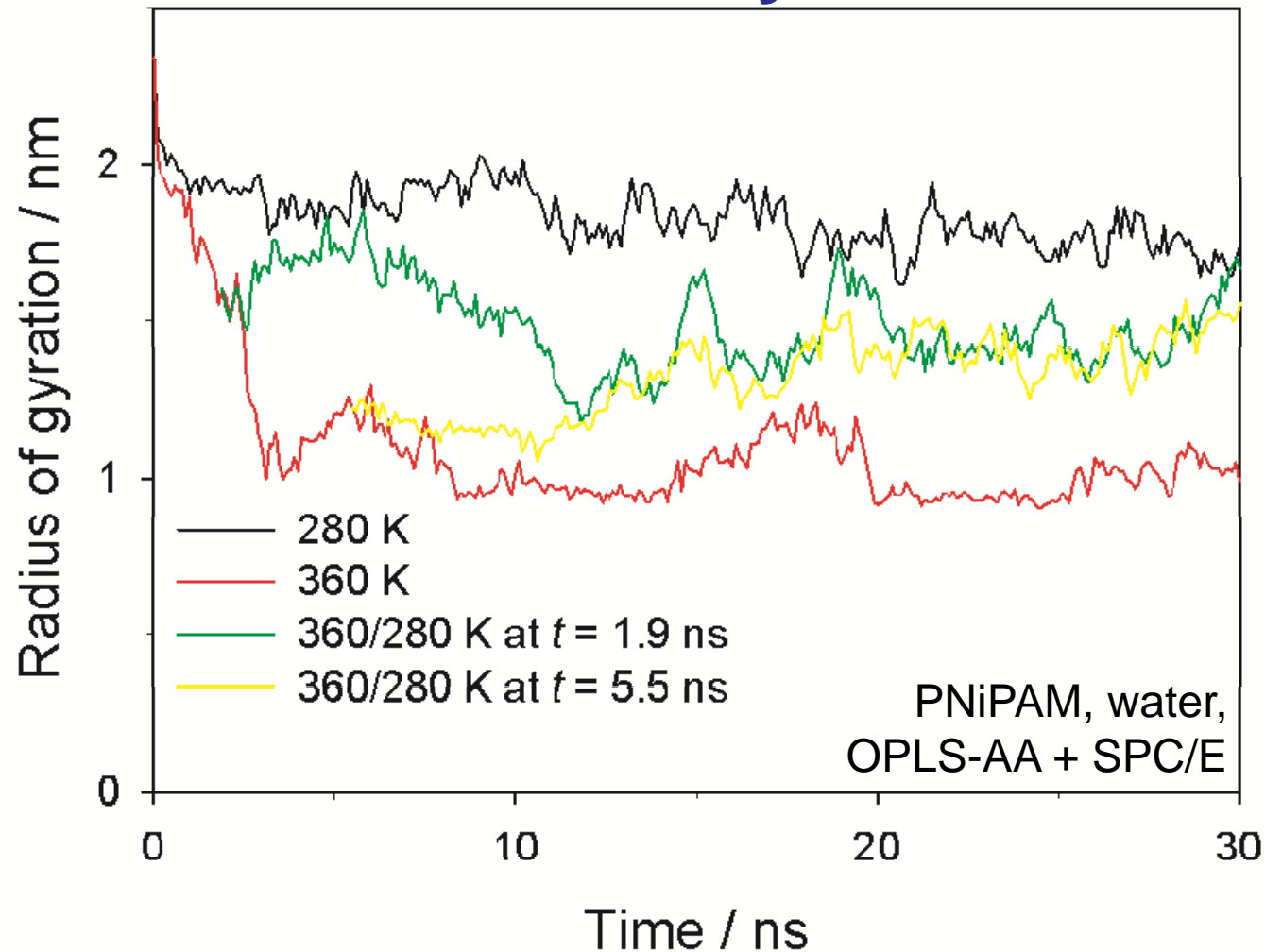


Temperature Dependence of Single Chain Radius of Gyration



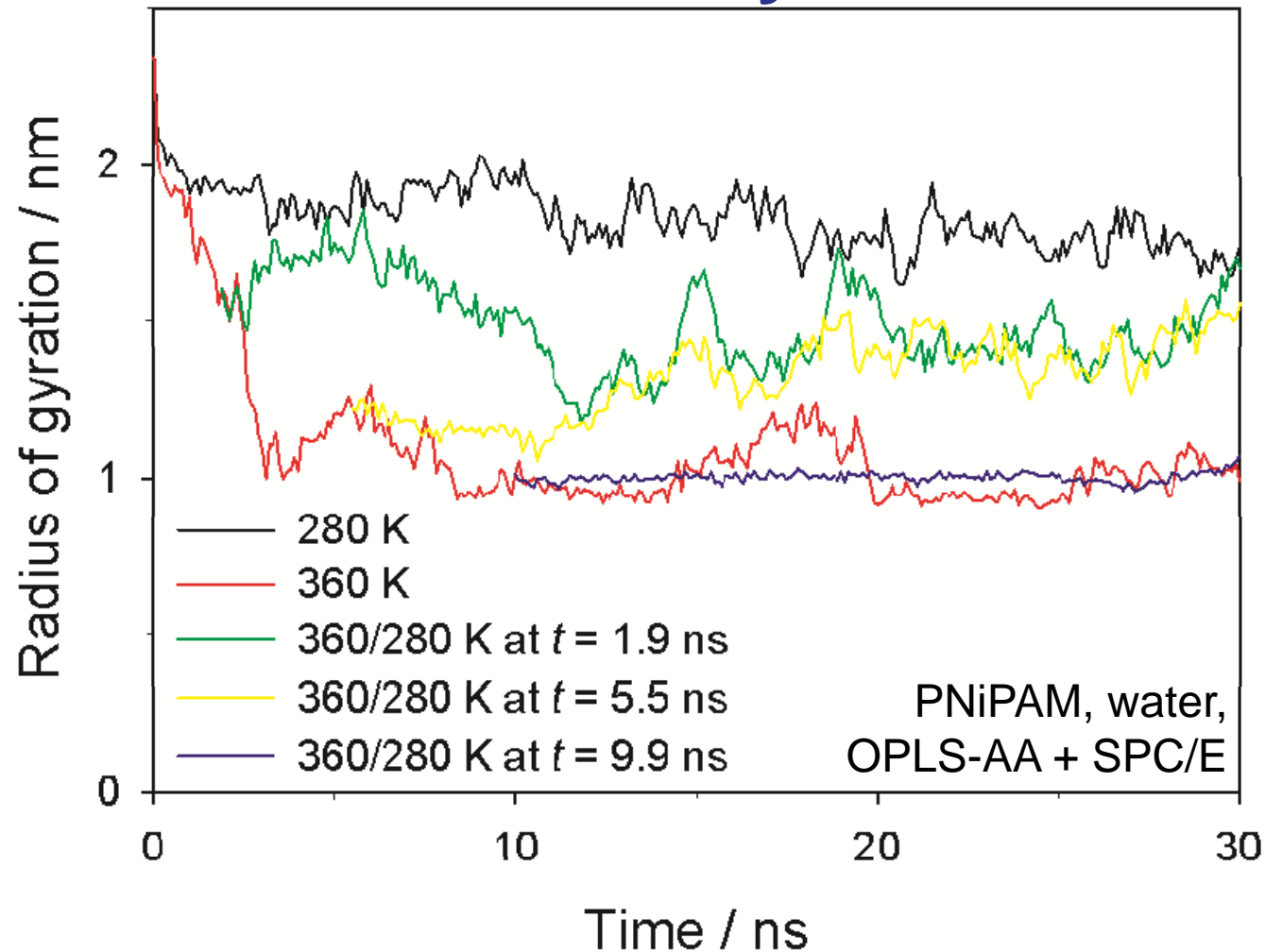


Temperature Dependence of Single Chain Radius of Gyration





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	+	+

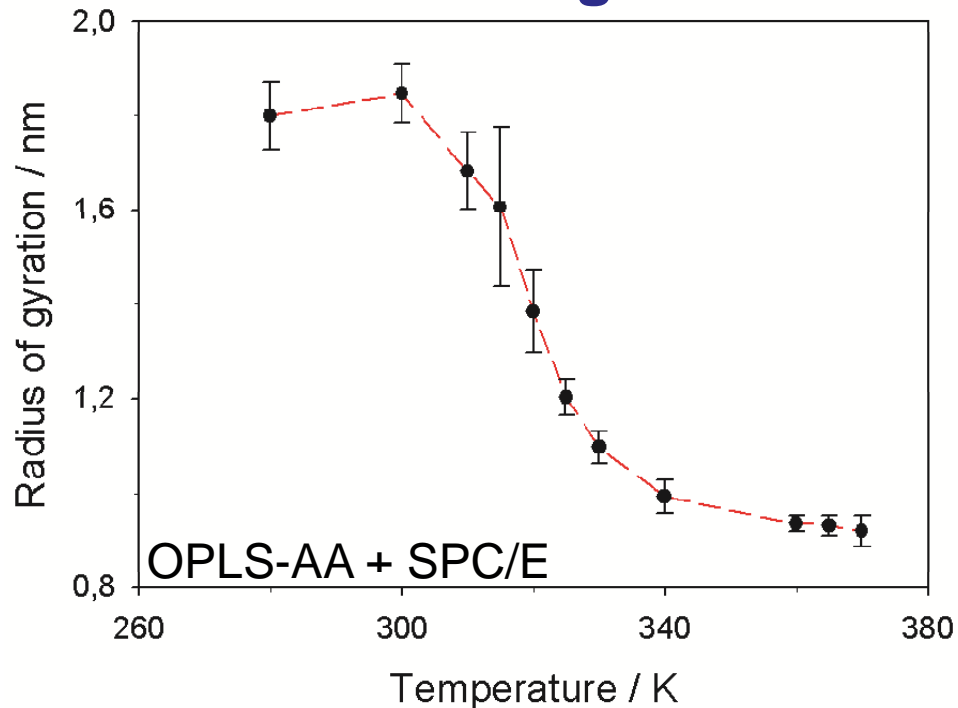
Legend

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

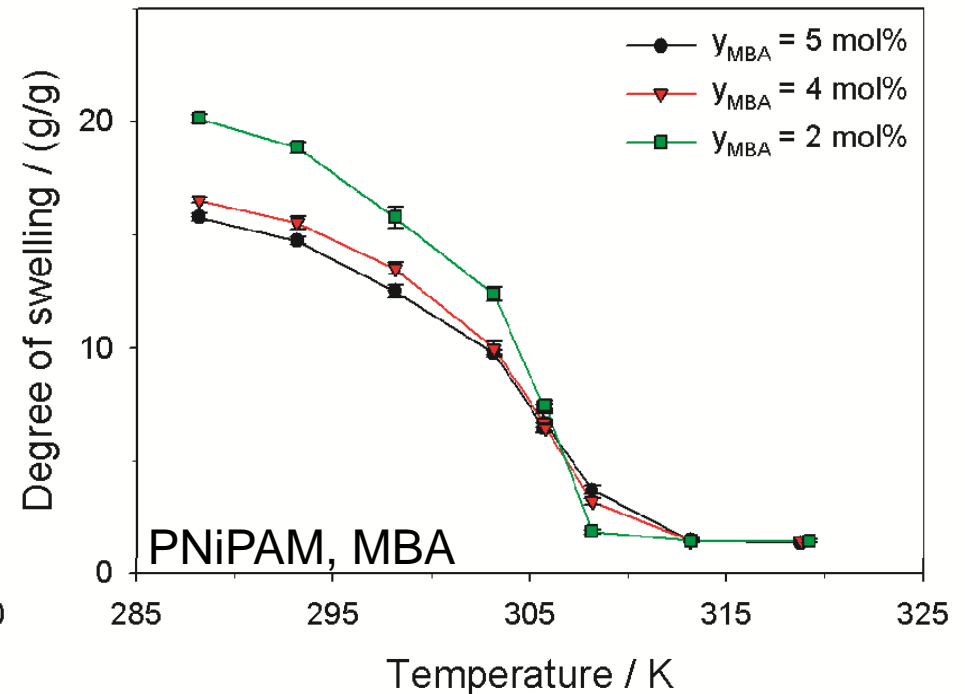


Temperature Dependence of the Swelling of PNiPAM in Water

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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