Experimental Report

Proposal: **TEST-2201** **Council:** 10/2012

Title: Chain dynamics in H-bonded double networks

This proposal is a new proposal

Researh Area:

Main proposer: **GOLD Barbara**

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Samples: Polyisopren in melt (deuterated ((C5D8)n) and pronated ((C5H8)n)
The backbones labelled with Urazole (C2H3N3O2) groups

Instrument	Req. Days	All. Days	From	To
IN15	6	6	05/07/2013	12/07/2013

Abstract:

-experimental report for experiment number TEST-2201-

Chain dynamics in H-bonded networks

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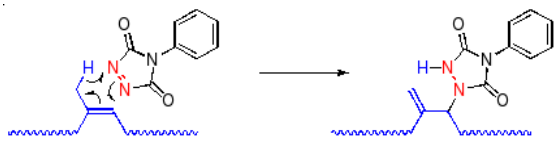
Nature uses a combination of dynamic hydrogen bonds and static covalent bonds to achieve elasticity in otherwise tough materials e.g. in the muscle protein TITIN [1]. If polymer chains are cross-linked by permanent chemical bonds, the system solidifies in disorder and becomes a rubber-like polymer network.
By modifying the polymer chains with dynamic hydrogen bonding groups, the mechanical strength of such networks can be enhanced. This allows the dissipation of high stresses by breaking the hydrogen bonds within the network. Subsequent re-combination leads to recovered stabilization.

As this mechanism is applicative for the development of new knowledge-based polymeric materials with advanced functionality like autonomous self-repair or innovative damage controllable materials, the main focus of this neutron spin echo experiment has been to investigate the influence of transient cross-links on the dynamics of entangled polyisopren chains.

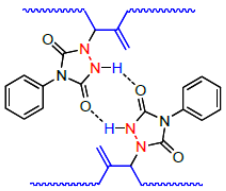
Synthesis of model system:

Adding hydrogen bonding urazole groups to protonated and deuterated polyisopren chains via Alder-ene reaction (fig. 1a) [2]. These functionalized chains are building a transient network via hydrogen-bonding (fig. 1b):

1a)



1b)



Samples:

Sample 1:
Blend of 85% protonated and 15% deuterated polyisoprene (M_w=100.000 g/mol),
no functionalization with transient cross-links

Sample 2:
Blend of 85% protonated and 15% deuterated polyisoprene (M_w=100.000 g/mol),
chains functionalized with urazole groups, functionalization degree: 1 mol%

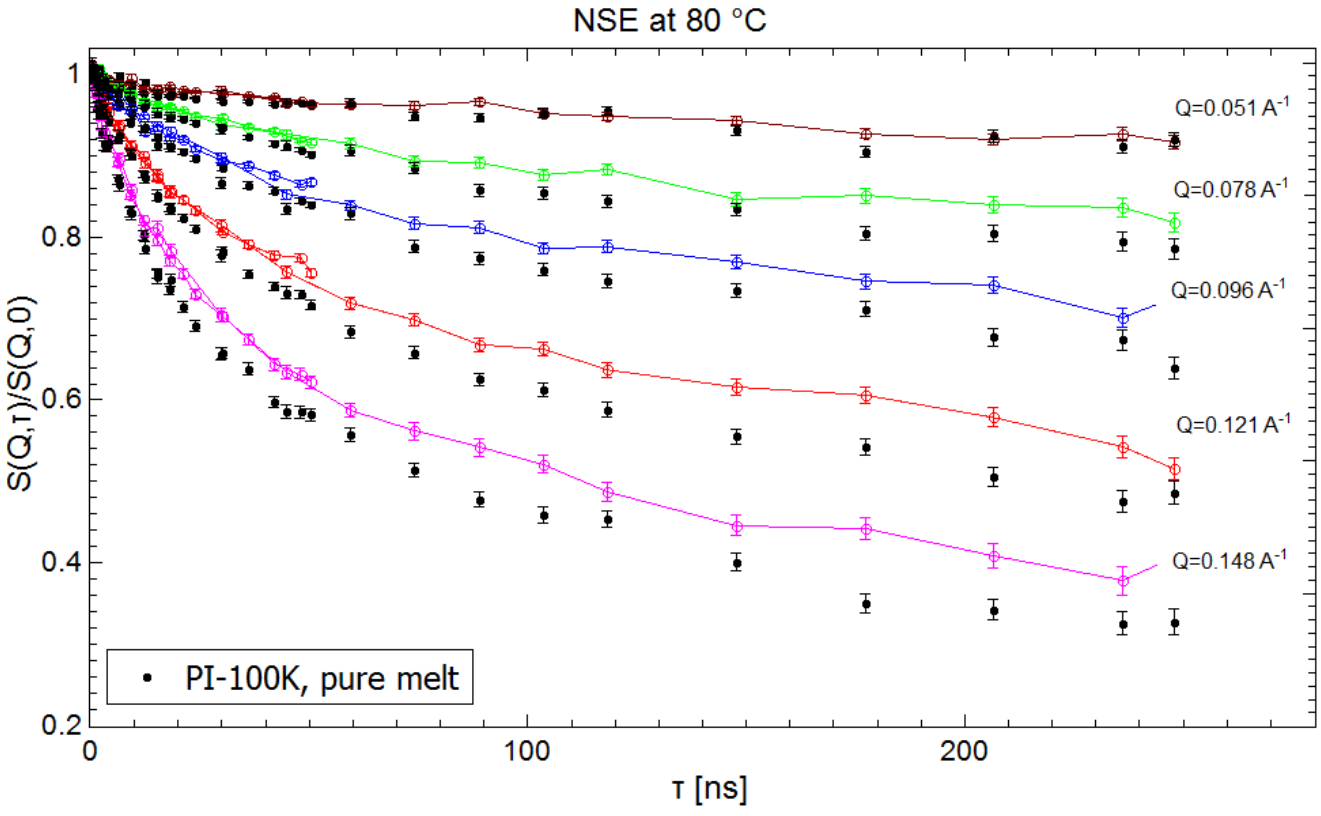
Sample 3:
Fully deuterated background sample, 100% deuterated polyisoprene (M_w=100.000 g/mol),
no functionalization with transient cross-links

Sample 4:
Fully deuterated background sample, 100% deuterated polyisoprene (M_w=100.000 g/mol), chains functionalized with urazole groups, functionalization degree: 1 mol%

Measured temperatures: 60°C, 80°C and 140°C
Measured Q-values: 0.051, 0.078, 0.096, 0.121, 0.148 and 0.3 $Å^{-1}$
Wavelengths used: 10 Å and 17 Å

Results:

The following graph shows a comparison of the background corrected NSE spectra between the unfunctionalized (sample 1, black dots) and functionalized (sample 2, coloured dots with lines) polyisoprene chains at 80°C. The Rouse region as well as the plateau region are affected by the additional transient cross-links, pointing to a higher segmental friction and a stronger topological confinement.



Data evaluation and modeling in progress. The short time regime will be modeled with the Rouse model, for the plateau regime the reptation model will be used.

References:
[1] Lee, E. H. et al., *Tertiary and secondary structure elasticity of a six-Ig titin chain*, Biophys J. 98(6): 1085–1095, 2010
[2] Stadler, R. et al., *Influence of hydrogen bonding on the properties of elastomers and elastomeric blends*, Makromol. Chem. 187: 1681-1690, 1986