

Experimental report

06/09/2016

Proposal: 9-11-1717

Council: 10/2014

Title: From nature to innovative self-healing materials:
The microscopic dynamics of self-healing polyisoprenenetworks

Research area: Physics

This proposal is a new proposal

Main proposer: Barbara GOLD

Experimental team: Barbara GOLD
Michael MONKENBUSCH

Local contacts: Peter FALUS

Samples: 50/50 HD polyisoprene blend functionalized with hydrogen bonding urazole groups. covalently cross-linked after functionalization

Instrument	Requested days	Allocated days	From	To
IN15	9	6	11/09/2015	17/09/2015

Abstract:

Double networks consisting of both permanent and transient cross-links are a new interesting class of materials which shows an extreme potential for applications with adaptive requirements. In the framework of a PhD thesis and an EU-funded FP-7 project (Self-Healing INnovative Elastomers SHINE) we recently mastered the synthesis, rheology and the SANS characterization of such systems under equilibrium. This proposal therefore aims at the microscopic investigation of the associated underlying dynamics at the length scales of the chains by means of NSE (IN15) in isotropic state. The first assessment of the non-equilibrium behaviour under strain by SANS (D11) has already been scheduled at D11 and will be measured in November 2014 (proposal 9-11-1673).

This proposal is a re-submission of the dynamic part of proposal 9-11-1673 as highly recommended by scientific council 2014-4.

experimental report for experiment 9-11-1717

**From nature to innovative self-healing materials:
The microscopic dynamics of self-healing polyisoprene networks**

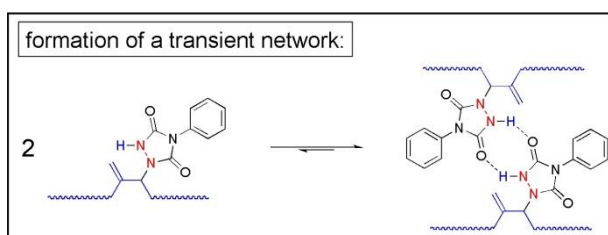
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Laboratory: JCNS-1 & ICS-1, FZ Jülich GmbH, 52428 Jülich

Instrument: IN 15

Aim of experiment: In the field of supramolecular polymers, dynamic temporary bonds are implemented inside common polymer melts or networks to create novel smart materials with self-healing and/or shape-memory properties. A prerequisite for the development of such tailor-made polymeric systems is a fundamental understanding of the underlying reassociation dynamics of the dynamic bonds and their influence on the intrinsic polymer dynamics. Therefore our recent studies aim to elucidate how the dynamics of entangled polyisoprene chains are influenced by backbone functionalization with reassociating urazole groups (see figure below).

Model system:



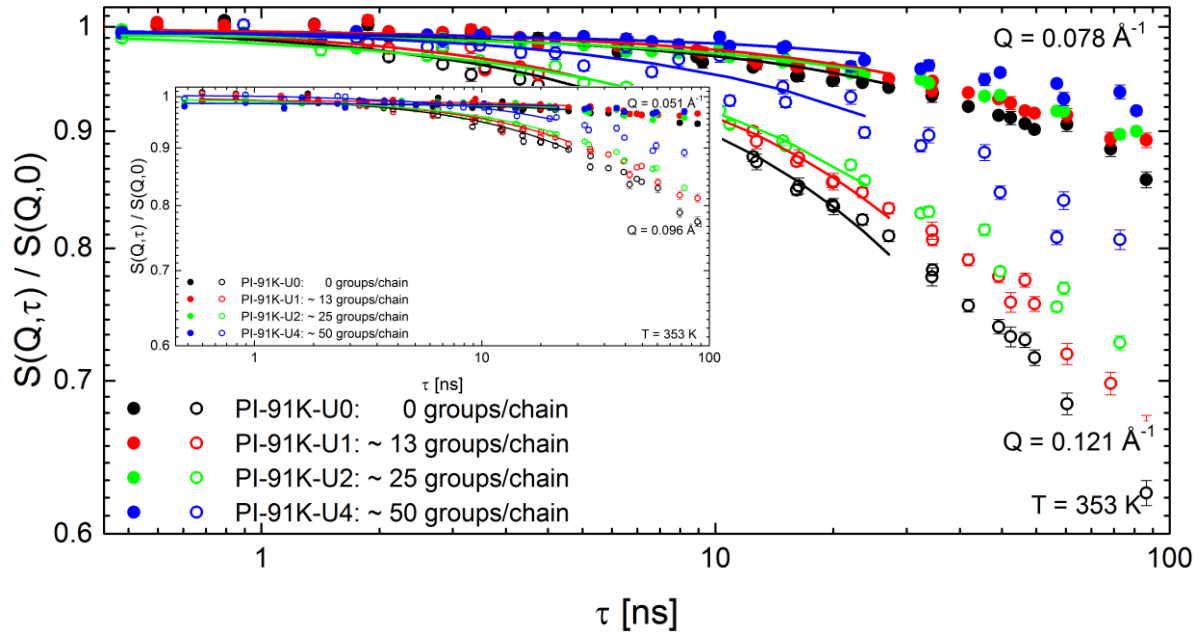
First neutron spin echo measurements have already been performed on IN15 (see proposal TEST-2201) for well entangled polyisoprene with a molecular weight of 91 kg/mol and a transient crosslink density of 1 mol% (13 groups/chain). The aim of the current proposal was the investigation of higher functionalization degrees (2 mol%, 25 groups/chain and 4 mol% 50 groups/chain) and their comparison with the results of the former experiment as well as with an unfunctionalized, pure linear, reference system.

Sample system:

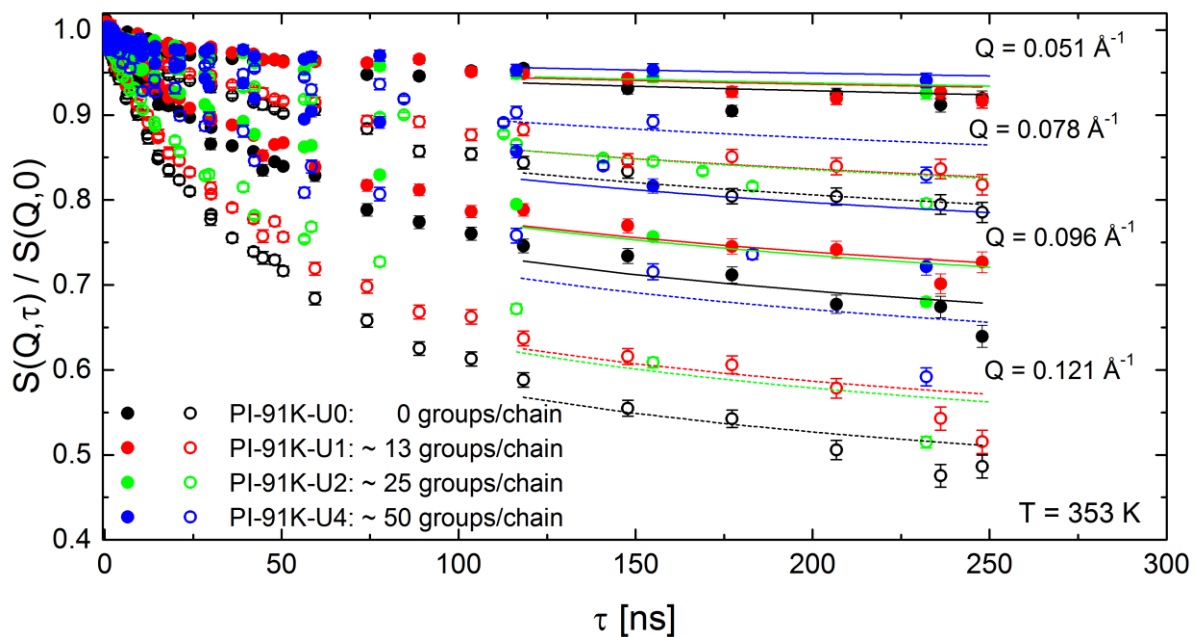
- PI (91 kg/mol), 0 mol% functional urazole groups (black symbols), PI-91K-U0
- PI (91 kg/mol), 1 mol% functional urazole groups (blue symbols), PI-91K-U1
- PI (91 kg/mol), 2 mol% functional urazole groups (red symbols), PI-91K-U2
- PI (91 kg/mol), 4 mol% functional urazole groups (green symbols), PI-91K-U4

Temperatures: 353 K and 413 K

Rouse regime - segmental friction: In the Rouse regime the reassociating groups lead to a slowing down of the dynamics, pointing to an increase of the segmental friction coefficient. The figure below shows the NSE data obtained for the model system (symbols) at $T = 353$ K (lines serve as a guide to the eye).



Plateau regime - topological confinement: In the plateau regime the reassociating groups lead to an increase of the plateau height, pointing i.e. to a slightly stronger topological confinement implemented by associated groups. The figure below shows the NSE data obtained for the model system (symbols) at $T = 353$ K (lines serve as a guide to the eye).



A quantitative description by modelling the data in terms of the Rouse (short times) and reptation (long times) model is currently underway.