Title:

Controlling ring mobility in slide-ring gels

Authors & affiliations:

Atul Kumar Sharma, Sina Ghiassinejed, Evelyne Van Ruymbeke, Charles-André Fustin

Université catholique de Louvain, Institute of Condensed Matter and Nanosciences (IMCN), Bio and Soft Matter division (BSMA)

Belgium

Abstract:

Polymer networks and gels show great interests for applications in various fields. Recently, the development of a new type of network, the slide-ring gels, has created new possibilities. In such gels, the polymer chains are held together by linked macrocycles, each having a polymer chain threaded into its cavity. The cross-links have thus the strength of covalent bonds (a ring must be broken to break the network) but are mobile since the rings can slide along the chains. Such sliding cross-links work as molecular pulleys and equalize internal tensions, leading to remarkable properties including huge swelling capacity, high stretchability, and high mechanical strength.

In this work, the main objective is to study the impact of the ring mobility on the gel properties. This is achieved by tuning rings-polymer chain interactions, while keeping all the other parameters (number of free rings, degree of crosslinking,…) constant.

We have synthesized slide-ring gels by two approaches exploiting urethane chemistry. In the first strategy, a polyrotaxane made of several macrocycles threaded on a polyurethane chain is synthesized, and the cross-linking is then induced by linking macrocycles with a bifunctional linker. In the second one-pot approach, a tetra-functional bis-pseudorotaxane is used as cross-linker in the copolymerization of poly(ethylene glycol) with a bis-isocyanide to yield the gels in one step. In both cases, the rotaxane motif is based on a palladium(II) complex between a tridendate and a monodendate ligands. The control over ring mobility is achieved by demetallation of these complexes, which frees the macrocycles, allowing them to move along the polymer chains. The slide-ring gels obtained by the two strategies have been characterized in detail by rotational shear rheometry, allowing us to systematically compare the properties of the two types of gels and to probe the influence of the ring mobility.