

Thermal and Mechanical Properties of New Main-Chain Liquid-Crystalline Elastomers

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Liquid-Crystalline Elastomers (LCEs) have the important combination of two properties: the entropic elasticity of polymer networks and the orientational anisotropic order of liquid crystal molecules. LCEs have only recently entered the actuator arena, with strains of up to 400% and characteristic stresses of up to 1 MPa.¹ The most unusual interesting properties found in such systems include spontaneous shape change at the phase transformation temperature, strain-induced transitions, dynamic-mechanical properties and soft elasticity.

In the early '80s, Aguilera and Ringsdorf² succeeded in synthesising Main-Chain Liquid-Crystalline Polymers (MCLCPs) based on three aromatic rings mesogenic units and oligosiloxanes as chain extenders. This kind of chemistry reduced the glass transition (T_g) and clearing temperatures (T_c) of those MCLCPs due to the flexibility of the siloxane spacer. Years later, Donnio, Wermter, and Finkelmann³ obtained the first Main-Chain Liquid-Crystalline Elastomers (MCLCEs) by crosslinking the liquid-crystalline polymer chains with a flexible siloxane-based crosslinker. In both studies, low degrees of polymerisation were obtained, from 6 to 9 repeating units, as could be noticed from the molecular weights of the polymers and from the high percentages of soluble content and crosslinker used in the networks.

New MCLCPs and MCLCEs have been synthesised⁴ based on reacting vinyl- or vinyloxy-terminated mesogens under hydrosilylation conditions with a flexible crosslinker. These main-chain systems showed smectic and nematic mesophases and their anisotropic properties have been mechanically and thermally analysed as function of the crosslinking density. Due to the suitable chemistry used in this work low crosslinking densities have been achieved (2.5 mol-%) with low soluble content (5%).

Uniaxial stress-strain deformations (Figure 1.a) were measured in order to study mechanical properties on these MCLCEs, and an exponential behaviour was observed for nematic elastomers; a pre-stress transformation region was also analysed for the smectic materials. In nematic elastomers, the mechanical

behaviour was correlated with the chemical constitution of each sample, showing a relation of direct proportionality between the degree of crosslinking and the elastic modulus, as opposed to the smectic elastomers. Thermoelastic experiments (Figure 1.b) showed thermal expansion differences ranging from 80% to 210%. For the first time, the degree of crosslinking could be adjusted and nematic or smectic MCLCEs with tuneable thermal and mechanical properties were achievable.

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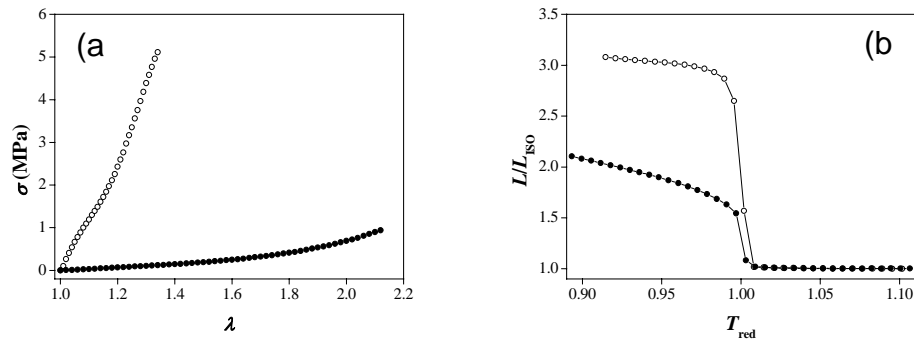


Figure 1. (a) Uniaxial stress-strain and (b) thermoelastic curves for a nematic MCLCE (●) and a smectic MCLCE (○) with the same crosslinking density (2.5 mol-%)