

Abstract

The incorporation of magnetic nanoparticles into polymer-based elastic matrices leads to soft composites with intrinsic magnetic properties. Typical for the occurring responsive material behavior in these hybrid composites is the coupling of the magnetic properties to the elasticity of the polymer network on mesoscopic length scales. This coupling is based on the mutual interactions between the embedded magnetic particles and the surrounding polymer matrix, and is influenced by the network's mesh size, the functionality of the matrix and the particle surface, as well as by the type of bond between the polymer network and the filler. By magnetic or mechanic stimulation, it is possible to induce anisotropic material properties in these hybrid composites. This leads to materials with direction-dependent macroscopic properties, which have enormous potential for applications as actuators or sensors.

In this work, the particle-matrix interactions in magnetic polymer composites are systematically analyzed. Starting with soft polymer-based ferrogels with comparably low strength of the mutual interaction, a model system of poly(acrylamide)-based hydrogels is employed, which shows a systematically varying mean mesh size in the size regime of superparamagnetic iron oxide nanoparticles. The magnetic nanoparticles are embedded in the model hydrogels with gradually decreasing mean mesh size. The increasing particle-matrix interactions lead to a gradual immobilization of the embedded nanoparticles in the meshes of the polymer network. Further, the resulting gradual inhibition of the *Brownian* relaxation mechanism is detected by a decrease of the heating efficiency of the embedded magnetic nanoparticles.

Stronger interactions between the embedded particles and the elastic matrix are found in particle-filled elastomers. Here, a novel material architecture is presented, which is formed by poly(dimethylsiloxane)-based elastomer networks that are exclusively crosslinked by highly functional inorganic nanoparticles. In comparison to conventionally crosslinked particle-filled elastomers, the swelling, thermal, magnetic, and mechanical properties of the novel hybrid materials are evaluated. The novel particle-crosslinked elastomers here show strongly particle-content depending properties as well as enormous strains at break of up to 1700 %.

For the hybrid particle-filled elastomers, spindle-type hematite particles are employed as particulate crosslinkers. These particles are thermally blocked and possess a permanent magnetic moment as well as an elongated shape, which makes them interesting candidates for the analysis of orientational as well as magnetic anisotropy in the hybrid composites. Hence, the possibility to incorporate and tune spatial as well as magnetic anisotropy in the prepared novel hybrid materials is further evaluated. The anisotropy is investigated by angle-dependent magnetization measurements in addition to small angle X-ray scattering experiments depending on the strain. Here, a tunable anisotropy is found, which depends on the sample architecture, the magnetic field, the particle volume fraction, and the applied strain.

The results presented in this thesis contribute to a better understanding of the particle-matrix interactions in particle-filled polymeric materials. The analysis of the occurring direction-dependent material behavior provides substantial insight into the generation and modification of anisotropy in particle-filled polymer networks, which is fundamental for novel actuator or sensor materials.