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| | Experiment title: Structural reorganization of magnetic nanoparticles during stimulus-healing of thermoplastic rubbers | Experiment number: SC-5161 |
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Report:

Abstract: We have investigated the heating mechanism in industrially relevant multiblock copolymers filled with Fe-nanoparticles subjected to an oscillatory magnetic field that enables their healing in a contactless manner. While this procedure aims to extend the lifetime of various thermoplastic elastomers, repeated or prolonged stimulus healing is likely to modify their structure, mechanics and ability to heat, which must therefore be characterized in depth. In particular, our work sheds light on the physical origin of the secondary heating mechanism observed in soft systems subjected to magnetic hyperthermia, being here triggered by the copolymer chains dissociation. In spite of earlier observations, the origin of this additional heating remained unclear. By using both static and dynamic X-ray scattering methods (small angle X-ray scattering and X-ray photon correlation spectroscopy, respectively), we demonstrate that beyond magnetic hysteresis losses, the enormous drop of viscosity at the polymer melting temperature enables motion of nanoparticles that generates additional heat through friction. Besides, the application of the oscillatory magnetic field for a few minutes is found to magnetize the nanoparticles, which causes their alignment into dipolar chains and leads to nonmonotonic translational dynamics. By extrapolating these observations to rotational dynamics and the corresponding amount of heat generated through friction, we not only explain the activation of a secondary heating mechanism but we also rationalize the appearance of a maximum in temperature during induction heating.

Figure 1 presents the SAXS intensity measured from the neat matrix, the iron powder and a composite loaded with 5 vol.% in particles at rest. This static analysis enables to extract typical sizes and exponents that will then be used to quantify the structural changes upon magnetic field irradiation.

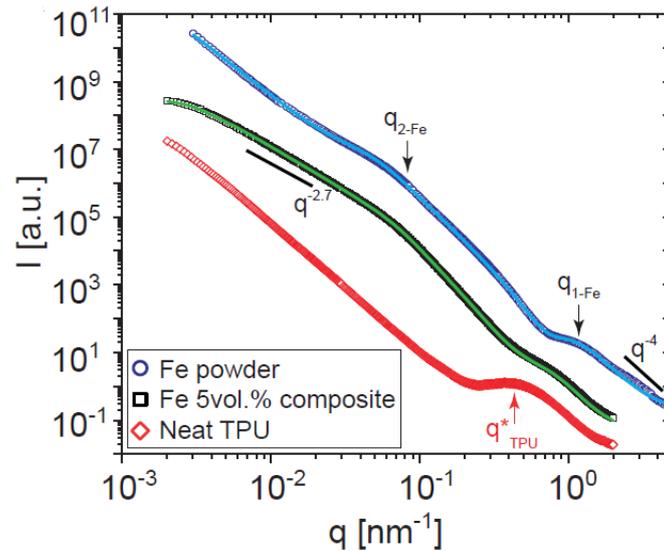


Figure 1 : SAXS intensity measured on the powder, the matrix and a composite at rest.

Above the melting point that is reached by magnetic hyperthermia, Figure 2 highlights the isotropy of the structure at short time whereas it reveals anisotropy at longer time. We added scanning electronic micrographs to further illustrate the structural change.

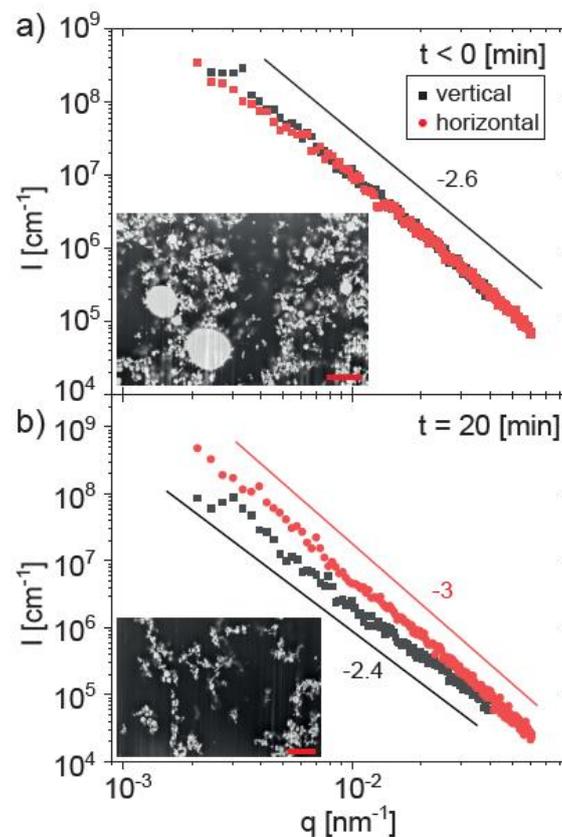


Figure 2 : Equatorial vs. Polar averaged SAXS intensity measured on the 5 vol.% composite above its melting point after a) 2 and b) 19 min of magnetic irradiation.

Figure 3 presents one of the XPCS experiment we performed on the composite loaded with 5 vol.%. In this case, XPCS data was acquired at short time above the melting point that was reached by magnetic hyperthermia. These data were then compared with analog data (i) recorded at the same temperature but without magnetic field (convective heating) and (ii) with magnetic field but longer irradiation time (not shown).

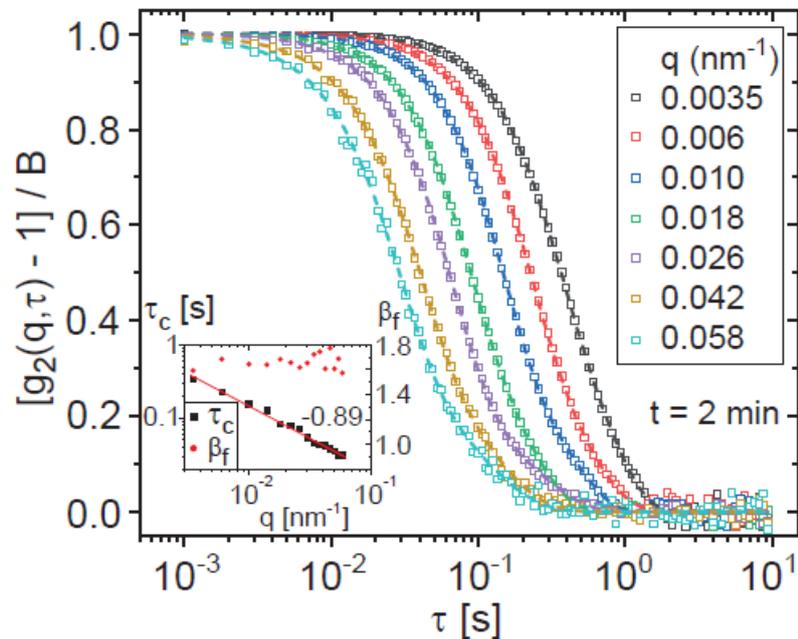


Figure 3 : Normalized XPCS autocorrelation functions obtained at various scattering vector two minutes after the melting of the composite containing 5 vol.% in particles. Dashed lines are fit with a KWW function, characteristic times and exponents are reported in the inset.

We present in Figure 4 a photograph and a schematic representation of the experimental setup

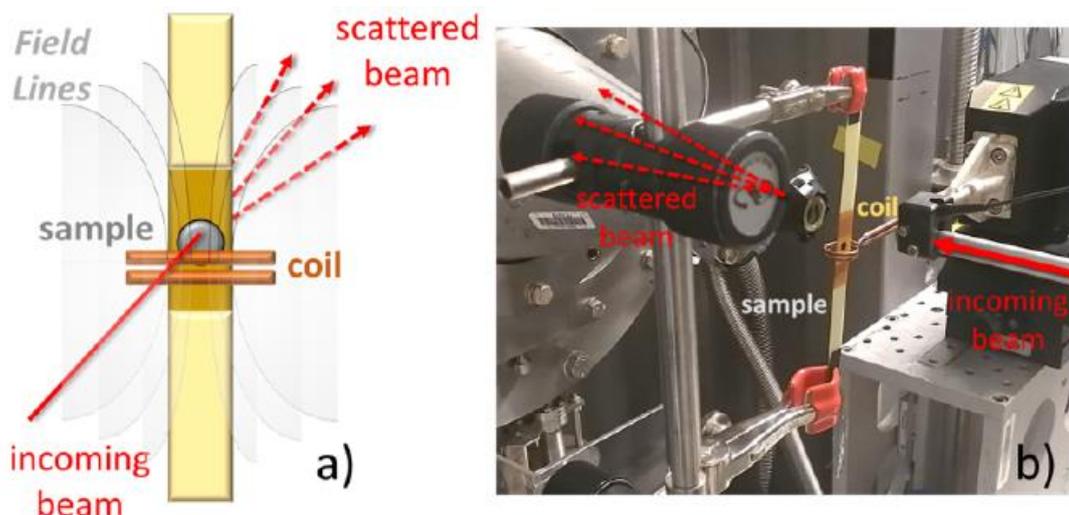


Figure 4 : Experimental setup. a) schematic representation. b) photograph on the ID02 beamline