The Mechanoresponsive Nature of Metallosupramolecular Polymers

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Abstract

The design principles of natural materials are frequently exploited to furnish artificial materials with tailored properties such as a responsiveness to mechanical stimuli. In these mechanoresponsive materials, a macroscopic mechanical force is translated into a chemical change on the molecular level. Metallosupramolecular polymers are attractive candidates as mechanoresponsive materials, due to their reversible assembly into dynamic structures and the tunable interaction strength of the binding motif that can be adjusted through a variation of the metal or ligand. We aim to prepare mechanically responsive materials based on metallosupramolecular polymers and investigate their behavior under stress from the macroscopic down to the molecular level.

Thus, polymeric networks were made by the coordination of metal ions to a telechelic poly(ethylene-co-butylene) that was end-capped with methylbenzimidazolyl pyridine ligands.^{2,3} Europium- and terbium-based supramolecular polymer films were successfully welded by ultrasonication as a means of mechanical stimulation and the interface of joined pieces was investigated by energy-dispersive X-ray spectroscopy to elucidate the diffusion of metals (Figure 1A). Moreover, telechelic polymers carrying ligands with extended were prepared to allow better analysis of the (dis)assembly of the metal-ligand complexes upon application of mechanical forces. Upon addition of europium the bright blue fluorescence of the unbound ligands is replaced by the red fluorescence of the europium-based metal-ligand complexes in solution as well as the solid state (Figure 1B). Application of ultrasound to solutions of the metallosupramolecular polymers resulted in a temporary decrease of the europium fluorescence, indicating the transient disassembly of the metal-ligand complexes. Investigating how these supramolecular polymers respond to mechanical stimuli in solution and in the solid state is envisioned to provide a detailed understanding of their mechanochemistry.

References

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Figure

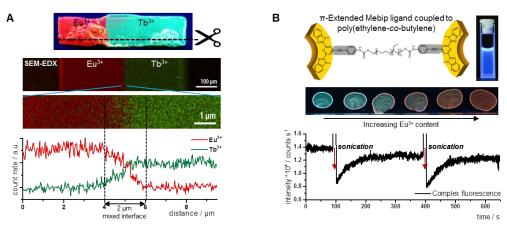


Figure 1: (A) Mechanochemical welding of europium- and terbium-lap joints and investigation of the welded interface. (B) Fluorescence of a π -extended ligand in solution, the solid state, and upon application of ultrasound.