The α-Relaxation as a Novel Shape Memory Switch

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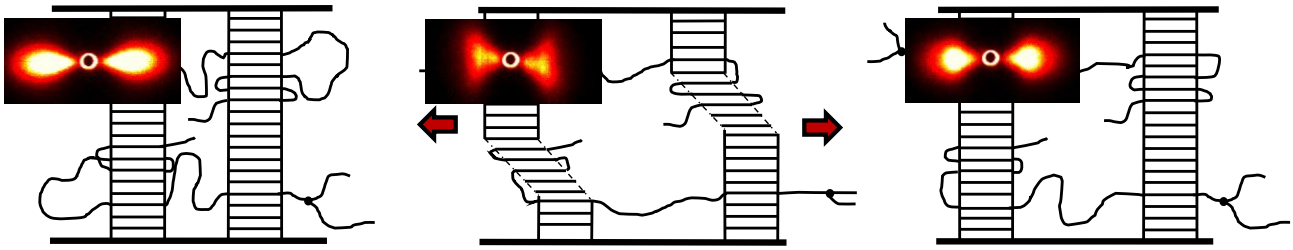
Thermally triggerable shape memory polymers (SMPs) typically exploit the glass transition temperature (T_g) or melting temperature (T_m) as switching mechanisms to transition from one shape to another. However, recent studies on cross-linked polyethylene networks have demonstrated that the α-relaxation temperature (T_α) can also serve as a novel trigger for cross-linkable semi-crystalline thermoplastics. T_α enables the migration of polymer chains through lamellar crystals, allowing for shape switching that is accompanied by a reversible change in morphology.

Initial investigations using cross-linked low-density polyethylene (xLDPE) showed that crystallizing the polymer under strain fixes it in an intermediate shape. Subsequent post-stretching at a temperature between T_α and T_m, followed by cooling, programs a temporary shape. Reheating to this intermediate temperature initiates the retraction back to the intermediate shape. Because the T_α of xLDPE partially overlaps with its broad melting range, the precise contribution of partial crystal melting to this effect could not be fully ruled out. [1]



To isolate the effect of the α-relaxation, cross-linked high-density polyethylene (xHDPE) was investigated. The thermal transitions of xHDPE offer a distinct separation, where T_α is well separated from the onset of melting. The xHDPE networks demonstrated reversible switching between temporary and intermediate shapes exclusively at temperatures above T_α and below T_m. This establishes that the α-relaxation alone drives the shape recovery without any melting, confirming it as a genuine switching mechanism in semi-crystalline SMPs. Furthermore, it enables the complete recovery of plastically deformed morphologies in the solid state, without the need for crystal melting. This concept opens up new possibilities for the design of advanced multi-shape memory materials within a single semi-crystalline network. [2]

[1] M. Maricanov, R. Becker, R. D. L. Jerusalem, J. C. Tiller, and F. Katzenberg, *J. Appl. Polym. Sci.* **141** (2024) e56241
[2] M. Maricanov, R. D. L. Jerusalem, C. Meykranz, L. Schultz, J. C. Tiller, and F. Katzenberg, *Polymer* **341** (2025) 129287