Micro-mechanical constitutive formulation of strain-induced crystallization in soft rubber-like materials

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Strain-induced crystallization (SIC) is one of the processes in which external mechanical loading can influences the internal micro-structure of the material. In this process, under large amounts of stretch, small crystalline phases nucleate inside amorphous rubber materials. This formation of crystals modifies the chemical and mechanical properties of polymer and leads to interesting phenomena such as the toughening effect of SIC around a crack tip [1].

We developed a micro-mechanically based constitutive model to simulate the combined mechanical deformation and phase transition in rubber using finite element method. The model describes the relationship between the measurable macroscopic effects and the underlying microscopic processes of the polymer chains. Our model establishes the link between the continuum mechanics description of deformation and its microscopic counterpart using the Maximal Advance Path Constraint model [2]. For each orientation of the chains, we use the stretch of the chains and its state of crystallinity to determine the chain's axial force and its rate of crystallization. Any standard chain model for SIC such as those in [3,4] can be used within our constitutive formulation. Another aspect of our SIC model is the macroscopic description of crystallinity, which includes the total of crystallinity as well as the orientation of the crystals and their distribution in various orientations. The information about the microscopic state of crystallinity of all the chains is represented by a small number of variables that can be measured in experiments. This set of macroscopic crystallinity variables allows us to study the anisotropic effects of SIC in rubber and the realignment of crystals under non-proportional loads. Using the rate of crystallization of the chains, obtained from the chain formulation, we compute the evolution of the macroscopic crystallinity parameter by performing a homogenization algorithm.

Our model shows quantitative agreement with the uniaxial loading and unloading experiments in both stress response and the evolution of crystallinity in the material. In addition to uniaxial tests, a 3D simulation is performed to study the effect of SIC in th vicinity of cracks. The simulation confirms the formation of crystals close to the crack tip and agrees with existing experimental evidence.

References

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