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POLYMER MODELLING: FROM MACROSCOPIC HYPERELASTICITY TO STRAIN INDUCED CRYSTALLISATION

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The purpose of the talk is to review the numerical modelling of the large deformation of structures made of soft materials and to outline present directions of research. The original models in isotropic finite elasticity consider the simplest case where stresses derive from an isotropic energy density function of the invariants of Cauchy Green strain tensor or of its eigenvalues. This has been extended to anisotropic hyperelasticity with the help of additional invariants and to more general constitutive behavior such as plasticity, viscoelastic or damage. In most cases, such models are based on a multiplicative split of the deformation gradient, and evolution laws governing the additional internal variables are obtained by postulating a specific form of dissipation within the material [5], [8]. From a numerical perspective, these internal variables can be eliminated from the tangent problem, reducing the general case to a succession of hyperelastic problems. This phenomenological approach is now used in most models, with different levels of complexity to take into account clusterisation, ageing, crystallization and so on.

To overcome the high level of arbitration inherent to the above phenomenological approach, it has been tried from the early days to relate the energy densities at the continuum level with the physically motivated free energy of polymer chains. The difficulty is to pass from one chain to a network of *cross-linked* chains, and this requires some constitutive assumption. Treloar did assume that the network deforms locally in an affine manner according to the macroscopic strain gradient. The resulting energy density and stresses are in good agreement with experiments in moderate deformation but are overestimated in large strains. To relax the affine assumption, a geometric response of a fictitious representative volume element of polymer can be postulated as in the eight-chain model of [1]. A more recent "variational" approach is to replace the affine assumption by a minimization principle introducing the macroscopic deformation either as a maximal path constraint as in [7] or through a microscopic boundary condition as in [3].

The Treloar idea of getting down to a lower scale has also successfully inspired different authors such as in [6] or [2] in their development of constitutive laws able to take into account evolutive anisotropic damage. Their micro-macro approach superposes a continuous distribution of one-dimensional independent stress-strain relations over the orientation space. We will illustrate this "microsphere approach" by a recent development done in [4] for strain induced crystallization. At the individual chain level, crystallization is described by a relative frozen length $\lambda\chi$ which enters the representative chain energy. The evolution law governing

the time evolution of this length is based on a thermodynamical argument controlling the force (free energy derivative) associated to $\lambda\chi$. As in plasticity, crystallization is authorized only if the force is equal to a given threshold. The extension of this 1D model to a full 3D setting is based on the micro-sphere framework through an affine network deformation assumption. This leads to an evolutive anisotropic model which is tractable and which has been numerically implemented in the open source finite element software FEBIO.

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