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A three-dimensional network model for rubber elasticity: The effect of local entanglements constraints

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ABSTRACT

We present a micro-mechanical model based on the network theory for the description of the elastic response of rubber-like materials at large strains. The material microstructure is characterized by chain-like macromolecules linked together at certain points; therefore an irregular three-dimensional network is formed. The material behaviour at the micro-level is usually described by means of statistical mechanics. Using certain assumptions for the certain distributions, one arrives at a continuum mechanical model of finite elasticity. However, the macromolecules interactions are neglected usually in these approaches. In the present contribution, we propose to add the effect of the interactions between chains of the cross-linked network. Following Arruda and Boyce (1993, 2000) [31,2], a cubic unit cell is defined where the entanglements fluctuations are localised in the corners of the cubic sub unit cell. These entanglements are linked by chains which ensure the interactions between the chains of idealized network (without interactions). These interactions can be represented by chains which are located in the principal directions of the cubic sub unit cell in undeformed state. We assume the probability densities which describe the free chain response of idealized network, and, the chain of constraints networks are independent. Then, the free-energy of the entire network is obtained by adding the free-energies of the free idealized (without interactions) and constraints (due to the chains interactions) networks. The constraint network reduces to four of the three-chain model of James and Guth (1943) [4] in undeformed state. Therefore, the free-energy of constraint network is obtained using the standard three-chain model, and, the free-energy of the free idealized network is constructed by means of the eight-chain model. The constitutive model involves five physical material parameters, namely, the shear modulus at small strains (μ_0), the numbers of links that form the macromolecular chain of the eight-chain, and three-chain models (N_8, N_3) respectively, a micro–macro variable K_i, and, non-dimensional parameters (η, ρ). In order to determine the material parameters, the Langevin function in the single chain configuration is replaced by its first order Padé approximant [see, Cohen (1991) [5]; Perrin (2000) [6]], and, the material parameters are identified. The excellent predictive performance of the proposed model is shown by comparative to various available experimental data of homogeneous tests. However, the present model requires a validation because the relationship between the micro and macro levels needs to be clarified. Indeed, the identification of the physical parameters (μ_f, μ_c, N_8, N_3) from experimental results data at micro is hoped in order to simulate the macroscopic (i.e. bulk) behaviour of the material.

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1. Introduction

The rubber-like materials are widely used in different industrial areas like rubber tires, engine and door seals, isolations or building bearings. Use of numerical simulations (finite element codes for example) is more and more used to design these complex mechanical structures. Even if many strain-energy functions are implemented in these codes, it can be difficult to get the relevant material parameters to make them run. On the other hand, the constitutive equation derived from these different strain-energy functions often fails to predict the response behaviour of rubbers for complex loadings in large strains, i.e., biaxial tension or coupled tension and shear for example. Our goal is to obtain a reliable three-dimensional constitutive equation which can be able to simulate the complex deformation modes of rubbers.

The hyper-elastic behaviour can be described with an elastic strain-energy function W from which the stress–strain relation derives. Such a function can be determined from purely phenomenological or a micro-mechanically based model (see Boyce and Arruda [2] for details). The phenomenological approach uses the invariants or principal stretches as variables to build the energy function. These formulations have generally a polynomial structures as functions of strain invariants of Cauchy-Green tensor (Mooney [7], Rivlin [8,9], Rivlin and Saunders [10], Gent and Thomas [11], Hart-Smith [12], Yeoh [13], Criscione et al. [14] and others) or the principal stretch ratios $(\lambda_i)_{i=1,2,3}$ (Valanis and Landel [15], Ogden [16–18], Béchir et al. [19] and others). One disadvantage of these models is the important number of material parameters which are not physically related, and the identification which demands experimental data of many independent loadings (for example, the data of the simple tension and equi-biaxial tension as in Chevalier et al. [20]). Ideally, these parameters should be obtainable from a small number of experiments, one if possible. In the Valanis–Landel approach [15], the shear modulus, μ_0 of the material can be determined by experimental results data of pure shear loading or simple tension test (see Peng and Landel [21]) in the domain of elongation between 1 and 2.5. Recently, Criscione et al. [14] developed a new approach which based on the Hencky strain tensor (i.e. logarithmic invariants). These invariants lead to models which can be fitted by experimental results data of pure shear test. We note that some parameters of these models have not physical significance. One concludes that most models are not able to represent the behaviour of rubber-like materials in various deformation states with only a small number of physically based parameters identified on a single test.

Besides these purely phenomenological models, micro-mechanically based idealized network models have also been proposed. The description of rubber elasticity by molecular network models has received considerable attention: Treloar [22], Erman and Mark [23] or Erman [24], give an excellent comprehension treatment of the subject. The theoretical development of these models is based on the statistical treatment of network of non-Gaussian chains. The microstructure of rubber is assumed to be constituted of long molecular-chains randomly distributed in space. A single chain which consists of several segments containing monomers, is defined by two linkages which are considered as chemically or physically entangled points of molecular chains. The physical links are, in general, not permanent and may change depending on strain. Chemical links are permanent and preserve the entanglement situation. The decrease in the number of entangled points due to deformation causes an increase in the average number of segments N in a single chain, enhances extensibility and reduces the stiffness of the material. It is important noting that this softening plays an important role in the manifestation of the hysteresis during cyclic strain of rubber as shown in Marckmann et al. [25]. Starting from the free energy of a single chain, it is possible to compute the total free energy of the network composing the microstructure of the rubber-like materials. Derivation of the free energy of a single chain necessitates the introduction of a distribution function for the molecular-chain end-to-end distance r. This distribution function must be non-Gaussian to take into account the finite extensibility of molecular-chains: the most widely used and simplest non-Gaussian probability distribution is due to Kunh and Grün [26]. Several affine molecular-chain network models have been proposed and employed for the evaluation of the deformation behaviours of rubber and amorphous polymer (Treloar [22], Treloar and Riding [27], Wu and Van der Giessen [28,29]). However, these models cannot be integrated analytically and must be handled numerically. This difficulty can be overcome on using the discrete models which are proposed by (James and Guth [4] three-chain model, Flory and Rehner [30] four-chain model, Arruda and Boyce [31] eigth-chain model). The three-chain and the four-chain are not isotropic, while the eight-chain model is quasi-isotropic. This last model is mathematically simpler than the others and it compares favourably with experiments; we note that all these models fail to predict the biaxial extension data. It is found that the three-chain model overestimate and the eight-chain model underestimate the mechanical behaviour of the full-network model.

To approach the full network model, Wu and Van der Giessen [28,29] proposed a phenomenological model which is a linear combination of the three-chain and eight-chain models, the weight of each model is adjusted by numerical simulation. Elias-Zúñiga and Beatty [3] improved this phenomenological composite model by taking into account the structures of three-chain and eight-chain models: N_3 and N_8 are respectively the numbers of monomers in a molecular-chain of the network in the three-chain and eight-chain models. The advantage of the network models is the small number of material parameters which are not related to physical considerations. For example, the three-chain model or the eight-chain models have two material parameters: the shear modulus at small strains μ_0, and the number of monomers N in a single molecular-chain. This last parameter depends on the deformation state (Arruda and Boyce [31]). Because, the physical links may suffer breakdown during the thermo dynamical process, molecular dynamics simulation suggests also a change in the entanglements situation. To take into account the change in the number of entangled points, namely, in the number of segments N, depending on the temperature change and deformation during the orientation process of rubber, a non-affine molecular-chain network theory