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Time to failure prediction in rubber components subjected to thermal ageing: A combined approach based upon the intrinsic defect concept and the fracture mechanics



MATERIAL

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ABSTRACT

In this contribution, we attempt to derive a tool allowing the prediction of the stretch ratio at failure in rubber components subjected to thermal ageing. To achieve this goal, the main idea is to combine the fracture mechanics approach and the intrinsic defect concept. Using an accelerated ageing procedure for an Ethylene-Propylene-Diene Monomer (EPDM), it is first shown that the average molar mass of the elastically active chains (i.e. between crosslinks) can be used as the main indicator of the macromolecular network degradation. By introducing the time-temperature equivalence principle, a shift factor obeying to an Arrhenius law is derived, and master curves are built as well for the average molar mass as for the ultimate mechanical properties. Fracture mechanics tests are also achieved and the square root dependence of the fracture energy with the average molar mass is pointed out. Moreover, it is shown that the mechanical response could be approximated by the phantom network theory, which allows to relate the strain energy density function to the average molar mass. Assuming that the fracture of a smooth specimen is the consequence of a virtual intrinsic defect whose the size can be easily estimated, the stretch ratio at break can be therefore computed for any thermal ageing condition. The estimated values are found in a very nice agreement with EPDM experimental data, making this approach a useful tool when designing rubber components for moderate to high temperature environments.

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1. Introduction

The use of rubber components is very widespread in many industrial domains as, for example, in ground or air transportation, medical equipment, electrical insulation, mechanical damping, etc. They can consequently be subjected to complex mechanical loadings in various

http://dx.doi.org/10.1016/j.mechmat.2014.07.015 0167-6636/© 2014 Elsevier Ltd. All rights reserved. environmental conditions. Because of the dependence of the mechanical properties either on the kind of loading or on the operating environment (light, humidity, temperature, oxygen, etc.), an optimal design of these components must account for the alteration of the mechanical properties due to ageing. Indeed, chemical and physical ageings are known to strongly modify the mechanical responses of such materials, but also the ultimate properties. Especially, in the presence of oxygen, the modification of the chemical structure of the elastomeric material is essentially attributed to the macromolecular chain scissions and



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crosslinking in addition to the break and reformation of crosslink nodes (Rajeev et al., 2003; Rivaton et al., 2005; Colin et al., 2007a; Tomer et al., 2007). These mechanisms induce the alteration of the mechanical properties (Clavreul, 1997; Assink et al., 2002; Colin et al., 2007a).

Chemical ageing of polymers is generally investigated by using analytical techniques allowing the measurement of their chemical, physical and mechanical properties (Colin et al., 2007a,b,c). Since ageing is a long time process, accelerated thermal ageing tests are often used to shorten their exposure duration and predict their operating life time. In fact, from these tests, results for lower temperatures are generally obtained using the time-temperature equivalence principle (Ferry, 1970; Treloar, 1971; Ha-Anh and Vu-Khanh, 2005; Gillen et al., 2006; Woo and Park, 2011).

The purpose of this contribution is the prediction of stretch ratio at failure in rubbers subjected to thermal ageing conditions. To achieve this goal, the main idea is to combine the fracture mechanics approach and the intrinsic defect concept. This combined approach¹ was successfully introduced to study the biaxial fracture of rubbers (Naït-Abdelaziz et al., 2012) and is extended here in order to propose a failure criterion accounting for degradation due to thermal ageing.

This paper is organised as follows. In Section 2, a general background on the main tools involved in this paper is presented. It mainly includes fracture mechanics of rubbers, large strain elastic constitutive models and time-temperature equivalence. Material and experimental procedure are detailed in Section 3. In Section 4, experimental results are reported. The predictive approach is presented in Section 5: estimations are compared to experimental data. Concluding remarks close the paper in Section 6.

2. A brief literature background

2.1. Fracture mechanics of rubbers

Since the pioneering work of Griffith (1921), fracture mechanics can generally be tackled via an energy balance which defines a parameter called the strain energy release rate G defined such as:

$$G = -\frac{\partial U}{\partial A} \tag{1}$$

where *U* is the potential energy and *A* is the crack area. At crack initiation, the strain energy release rate *G* takes a critical value generally called the fracture energy and noted G_c which is considered as an intrinsic material property. Based upon this definition, Rivlin and Thomas (1953) introduced the tearing energy T_c , equivalent to G_c , in the case of elastomers. From a simple analysis of the modification in the stored energy in a specimen containing an edge crack of length *a*, they expressed T_c in the following form:

$$T_c = G_c = 2k(\lambda_c)W_c a \tag{2}$$

where W_c is the critical strain energy density and k is a factor depending on the stretch ratio $\lambda = l/l_0$. The suffix c denotes the critical value corresponding to crack initiation.

The k factor depends on the specimen geometry. As an example, according to Greensmith (1963) in the case of Single Edge Notched Tension (SENT) specimens, this parameter is expressed as follows:

$$k(\lambda) = \frac{2.95 - 0.08(1 - \lambda)}{\sqrt{\lambda}} \tag{3}$$

This result, based upon experimental data, was confirmed by a finite element (FE) analysis by Lindley (1972). Let us note that the energy interpretation of the *J* integral introduced by Rice (1968) and Cherepanov (1968) is equivalent to the strain energy release rate *G* for elastic materials. Thus, the fracture energy can be also evaluated by using the *J* integral. The fracture mechanics properties are dependent on the sharpness of the notch (Trapper and Volokh, 2008; Volokh, 2013) and this influence decreases when large strain is involved since the crack radius increases to form a smooth notch (Naït-Abdelaziz et al., 2012).

Dealing with multiaxial fracture of rubbers, in the case of circular defects, Naït-Abdelaziz et al. (2012) have proposed a general form for *T* which accounts for the biaxiality ratio at large strain:

$$T = G = J = 2\lambda_{eq}^{\gamma} g(\lambda_{eq}) Wa \tag{4}$$

in which γ is a parameter depending on the biaxiality ratio ranging approximately from 1 (uniaxial tension) to 1.4 (equibiaxial tension) and λ_{eq} is the equivalent stretch ratio expressed as a function of the principal stretches:

$$\lambda_{eq} = \sqrt{\frac{\lambda_1^2 + \lambda_2^2 + \lambda_3^2}{3}} \tag{5}$$

in which λ_1 , λ_2 and λ_3 are the principal stretches.

The function g in Eq. (4) depends on the equivalent stretch ratio and can be fitted by the following equation:

$$g(\lambda_{eq}) = 0.255 + \frac{2.837}{(\lambda_{eq})^2} - \frac{2.888}{(\lambda_{eq})^4} + \frac{2.507}{(\lambda_{eq})^6}$$
(6)

2.2. Constitutive mechanical models

The basic features of the rubber stress–strain response are generally described by large strain elastic constitutive models which can be classified into two categories: the first one, physically-based, is issued from statistical mechanics theories (Treloar, 1943; James and Guth, 1943; Arruda and Boyce, 1993) and the second one, phenomenological-based, is issued from invariant-based and stretch-based continuum mechanics approaches (Rivlin, 1948; Ogden, 1984; Yeoh, 1990). The invariant-based Rivlin (1948) and stretch-based Ogden (1984) phenomenological models are probably the most popular and are expressed in terms of strain energy density (SED) functions, respectively, as follows:

¹ It is based upon the assumption that defects always exist in a given material and act as stress concentrators. Consequently, the material failure is the consequence of the growth of virtual cracks.

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