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Adhesion, friction and viscoelastic properties for non-aged and aged Styrene Butadiene rubber

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ABSTRACT

We study adhesion and friction between smooth glass, and fresh and aged Styrene Butadiene – Isoprene rubber blend (SBR-IR). The friction and adhesion are only slightly modified by the aging process, but elongation at break, and rubber toughness, are strongly reduced. We attribute this to changes in the crosslink density, a decrease in the filler matrix strength, and to the formation and growth of crack-like defects. The latter have a small influence on adhesion and friction, but a large effect on the elongation at break.

1. Introduction

Contact mechanics and adhesion are central topics in Tribology [1–3] with applications to tires, seals, human joints, pressure sensitive adhesives, granular matter, wiper blades and syringes, to name just a few. Contact mechanics for stationary elastic solids with randomly rough surfaces in the absence of adhesion is now well understood [4–10]. However, including adhesion and friction, the problem becomes much more complex [4,11–18], in particular for real materials like rubber with viscoelastic (and non-linear) properties [19–23].

In the present work we study the influence of aging on adhesion and friction between smooth glass surfaces and Styrene Butadiene – Isoprene rubber blend (SBR-IR). We show that adhesion and friction are nearly unchanged by the aging process, while the elongation at break is strongly reduced. We note that the strength of materials often is determined by a low concentration of "large" defects, which strongly reduce the elongation at break, but which have only a small influence on properties such as friction and adhesion at macroscopic scale, which are averages over large rubber surface or volume elements.

Aging of polymers such as rubber can due to many different chemical and mechanical processes, facilitated by the high stresses and temperatures which may prevail in sliding contacts, or by the influence of ozone or sun light. High temperatures in particular may accelerate oxidation and radical-activated chain scission processes. Those processes may be called chemical degradation and certainly will impact the properties of the material. In addition elastomers have a significantly larger configuration space than other materials like steel and plastics related to the entropic component of their free energy (see Fig. 1). Consequently the performance of an elastomer as it ages will be affected not only by chemical degragation (bond breaking and bond formation processes), but also from molecular rearrangements influencing the entropic processes which are essential to their elasticity [27,28]. This aspect and approach to rubber aging will be discussed more in detail and depth in a future article from these authors.

Several aging processes are thermally activated, and consequently their rate k depend on the temperature. In the simplest case the temperature dependency of the aging rate follows an Arrhenius rate equation:

$$k = A\exp\left(-\frac{E_a}{k_B T}\right)$$

where E_a is the activation energy of the reaction, A is the pre-factor, also called collisional factor, k_B is the Boltzmann constant and T is the absolute temperature. The activation energy E_a is usually determined empirically by fitting the Arrhenius rate equation to the temperature dependency of the rate of change, k, of some physical, age dependent, property.

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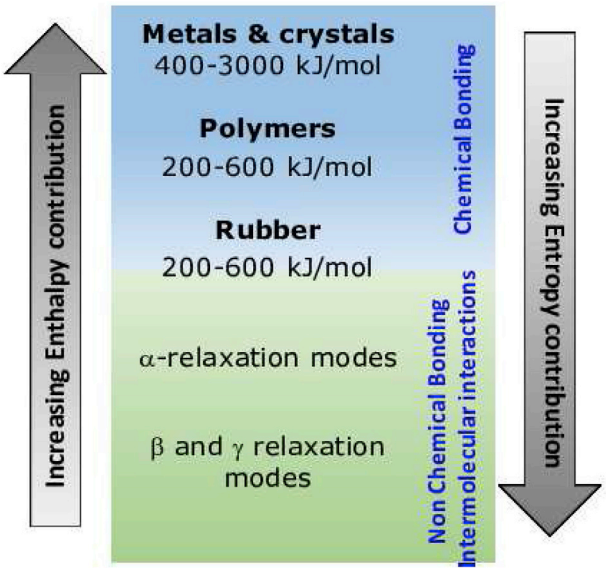

Fig. 1. In contrast to "hard" materials like metals, the mechanical properties of rubber materials are strongly influenced by entropic processes. Thus, slow time-dependent, thermally activated, molecular rearrangements will influence the mechanical properties of rubber (aging process).

This paper is organized as follows: In section 2 we briefly review the JKR theory, which are used to analyze the experimental data. In section 3 we present the experimental set-up and procedures used for the adhesion, friction and other measurements. In section 4 we present the experimental data with a brief discussion. The summary and conclusions are presented in section 5.

2. Short review of the JKR theory

The analysis of the experimental adhesion data is based on the JKR theory [24]. The contact region between a spherical probe (radius R) and a flat rubber surface is circular with the radius r. The interaction between the solids is described by the work of adhesion w, which is the energy per unit surface area to separate two flat surfaces from their equilibrium contact position to infinite separation. According to the JKR theory the relation between the force F and the radius r on the stable branch of the interaction curve is

$$r^3 = \frac{3RF_c}{4E^*}\left[\frac{F}{F_c} + 2 + 2\left(\frac{F}{F_c}+1\right)^{1/2}\right], \quad (1)$$

where $E^* = E/(1-\nu^2)$ (where E and ν are the rubber Young's modulus and Poisson ratio, respectively), and where

$$F_c = \frac{3\pi}{2} wR, \quad (2)$$

is the pull-off force. Thus for an elastic solid, if the ball is pulled by a soft spring (and neglecting inertia effects), at $F = -F_c$ the pull-off force abruptly drop to zero.

It is well known that the separation line $r = r(t)$ can be considered as a crack tip [25]. The work of adhesion w in general depends on the velocity $v_r = \dot{r}$ of the opening (during pull-off) or closing (during contact formation) crack. At finite crack velocity, for an opening crack w can be strongly enhanced, and for a closing crack strongly reduced, compared to the adiabatic (infinitely low crack tip velocity) value w_0. One contribution to the work of adhesion is derived from the viscoelastic energy dissipation in the vicinity of the crack tip (see Fig. 2). For an opening crack this will enhance w with a factor $1 + f(v_r, T)$, which depend on the crack tip velocity v_r and the temperature T. For a closing crack the corresponding reduction factor is approximately [26] $\approx 1/(1 + f(v_r, T))$.

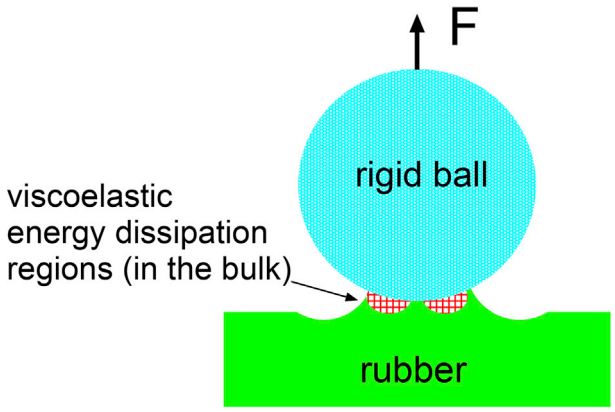

Fig. 2. A rigid ball pulled away from a viscoelastic solid. A part of the energy needed to remove the ball is derived from the viscoelastic energy dissipation inside the rubber close to the opening crack tip (red dashed region). (For interpretation of the references to colour in this figure legend, the reader is referred to the Web version of this article.)

Since the work of adhesion depends on the crack tip velocity $v_r = \dot{r}(t)$ we need to determine this quantity. We calculate v_r from the time dependency of $F(t)$ assuming that the JKR theory is valid. Thus using (1) we can obtain $r(t)$ from the measured $F(t)$. During pull-off the velocity v_r varies with time, but what is most important is the velocity at the point when the pull-off force is maximal; this is the crack tip velocity quoted in the experimental viscoelastic section (Sec. 4).

3. Experimental

Rubber aging–Plates of SBR-IR rubber (80:20) were aged in a convection oven at 80 ± 1°C for 4, 6, and 8 weeks. The non-aged and aged rubber samples were then used for the experimental procedures described below.

Adhesion–We have studied the adhesion interaction between spherical silica-glass balls and rubber. In the experiments we bring a glass ball with diameter $2R = 4$ cm into contact with a rubber substrate as shown in Fig. 3. It is positioned on a very accurate balance (analytical balance produced by Mettler Toledo, model MS104TS/00) which has a reproducibility of 0.1 mg or $\approx 1\ \mu$N. After zeroing the scale of the instrument we can measure the force on the substrate as a function of time which is directly transferred to a computer at a rate of 1 measurement point per second.

To move the glass ball up and down we have used an electric motor coiling up a nylon cord, which is attached to the glass ball. The pulling velocity as a function of time can be specified on a computer. In the experiments reported on below the glass ball is repeatedly moved up and

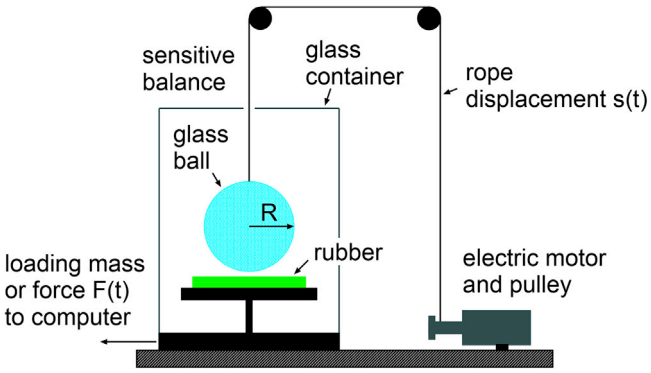

Fig. 3. The experimental set-up for measuring adhesion.

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