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Adhesion and heat build-up of rubber for energy-saved tyre

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Abstract. The objective of this study was to investigate the effect of crosslinking systems (conventional and efficient systems) on the contact mechanics and heat build-up of natural rubber (NR) tyre mimicking samples. The JKR (Johnson, Kendall and Roberts) experiment has been established to measure the contact mechanics in NR samples at different vulcanization systems. Work of adhesion obtained from JKR model is in good agreement with that calculated from contact angle measurement. From the contact mechanics point of view, the adsorption of the rubber to the glass slide surface plays a major role in the strain energy release rate. Higher rubber free chain samples (unvulcanized rubber) represent higher adhesion hysteresis and higher heat build-up compared to vulcanized rubbers whichever was the vulcanization system. Finally, rubber sample obtained using efficient vulcanization has the lowest adhesion hysteresis and heat build-up. These results suggest that the rubber with efficient vulcanization is preferable for the application of energy-saving tyres.

1. Introduction

Natural rubber (NR) was widely used as a viscoelastic material in many types of product [1-2]. Based on the global demand of energy-saved products, understanding of contact mechanics and vulcanization system of NR is very important. When a hemispherical rubber substrate contacts with another flat object, the adhesion forces acting across the interface tend to deform the solids and thus increase their contact area. At its simplest level, the experimental methodology involves bringing a deformable hemispherical object into contact with a flat substrate under controlled loads. Mechanical calibration of the load-deformation data using the equation yields the adhesion energy [3]. The Johnson-Kendall-Roberts (JKR) contact mechanics test is a simple test based on contact mechanics that has been more fully described elsewhere [4-5]. The JKR theory modifies the classical Hertz [6] result for the deformation of a sphere in contact with a flat surface to include the effect of interfacial energy or work of adhesion; the contact radius, a , is related to the applied load, P , by

$$a^3 = \frac{R}{K} \left[P + 3\pi WR + \sqrt{6\pi WRP + (3\pi WR)^2} \right] \quad (1)$$



where R is the radius of the undeformed sphere, K the elastic modulus and W the work of adhesion of the hemispherical polymer and the substrate. A typical JKR experiment is performed in three steps. The first stage is known as the loading process, which involves compressing the hemispherical elastomer against a flat surface under controlled loads. By determining the variation of the contact radius with applied load, it is possible to fit such data to equation (1) and to extract two independent properties K and W [4]. In the second stage, the elastomer remains in contact with the substrate for a fixed period of time. During this time interval, the evolution of physical and chemical interactions may lead to an enhancement in the adhesion energy. The final stage, or the unloading process, entails rapidly decompressing the hemispherical elastomer from the flat surface. One can then determine the strain energy release rate, G , necessary to maintain a certain radius of contact, a , between a flat surface and an elastic hemisphere of radius R under a contact force, P , by

$$G = \frac{\left(P - \frac{Ka^3}{R}\right)^2}{6\pi Ka^3} = \frac{(P - P_H)^2}{6\pi R P_H} \quad (2)$$

where $P_H = Ka^3/R$ is the Hertz contact pressure, the pressure that would be required to produce the measured a if there were no adhesive forces ($G = 0$). If equilibrium loading and unloading conditions are achieved, and if there is an absence of interface reconstruction, G should equal W , the equilibrium work of adhesion. In the case of two identical surfaces in contact, W is defined as twice the surface energy of that material [7]. The contact mechanics relates to the hysteresis of NR products, thus the lost energy of materials can be investigated [3,7]. The objective of this study is to investigate the effect of vulcanization system on the contact mechanics and vulcanization system of NR samples.

2. Experimental

Thai rubber block (STR 20) is used as a raw material for preparation of NR samples. The composition of rubber samples is as stated in table 1. Samples were prepared with internal mixer (type kneader) and two-roll mill.

Table 1. Formulas of rubber compound.

Formula (phr)	Control	Conventional Vulcanization (CV)	Efficient Vulcanization (EV)
NR (STR20)	100	100	100
ZnO	-	3	3
Stearic acid	-	2	2
TBBS	-	1	3
Sulphur	-	3	1
Total	100	109	109

Three types of hemispherical rubber compounds (control, CV and EV) were prepared in a hemispherical mould of 1 cm diameter using the procedure described elsewhere [3] for the study of contact mechanics using JKR experiment. The surface energies of the different rubbers and glass slide were evaluated from contact angle measurements using different liquid drops of known properties [2]. Tensile test is one of the most common tests to study mechanical properties of rubber. For the tensile test, rubber samples were cut in a dumbbell shape. The rate of testing crosshead speed was 500 mm/min at room temperature. Each material of the rubbers was tested at least three times. Crosslinking density of rubber networks is measured by its swelling, the number of effective network

chains per unit volume of rubber can be calculated according to the Flory-Rehner equation [8]. Heat build-up was measured by BF Goodrich Flexometer (Model II) following ASTM D623 method of measurement.

3. Results and discussion

Table 2 shows that NR compound of CV trends to have lower crosslinking density than that of EV in good agreement with the tensile strength from tensile test. On the other hand, NR compound of CV is better in elongation than that of EV. NR with conventional vulcanization (CV) represents long sulphur crosslinking bridge compare to NR with efficient vulcanization (EV), longer sulphur bridge better elongation. NR samples with both CV and EV clearly represent better mechanical properties (tensile strength and elongation at break) than unvulcanized NR (control).

Table 2. Properties of NR compounds.

Properties	Control	CV	EV
Crosslinking density (mole/m ³)	-	105 ± 3	110 ± 3
Tensile strength (MPa)	0.13 ± 0.01	23.95 ± 1.20	28.45 ± 1.42
Elongation at break (%)	248 ± 25	1826 ± 183	1383 ± 138

The surface energy of natural rubbers and glass slide can be determined using contact angle measurements. There are no significant differences of surface energy of all NR samples (below 50 mJ/m² means non-polar materials). For the glass slide, it represents as a polar material (above 50 mJ/m²). Concerning to the work of adhesion [2] between rubber samples and glass slide, there are no significant differences of work of adhesion from contact angle measurement: control/glass, CV/glass and EV/glass (table 3). Based on the contact mechanics test, a load and unload experiment was carried out to examine the contact mechanics of the hemispherical rubber [8]. The applied load (P) was increased stepwise to the final load and then released stepwise to the initial load. At each load, the radius of the contact zone (a) between the hemispherical rubber and glass slide was measured. Concerning to the relationship between a^3 and P for the loading experiment, typical results obtained during the loading experiment were fitted to equation (1) to extract work of adhesion (W). We found that work of adhesions obtained from JKR model are in good agreement with those calculated from contact angle measurement (table 3).

Table 3. Work of adhesion (W) of all NR samples.

Work of adhesion (mJ/m ²) ± 10%	Control/glass	CV/glass	EV/glass
Calculation from contact angle measurement	97.32	98.08	99.28
Extraction from JKR model	90.13	91.95	92.45

A typical JKR experiment is used in this study, it consists of bringing into contact two samples in the shape of a semi-spherical NR and a glass slide under the influence of an external load that increases until maximum load is reached and subsequently retracting them till the contact is broken. The adhesion hysteresis of PDMS networks has also been reported in the literature [4]. It is therefore of interest to investigate such adhesion hysteresis in our system because it should be related in some instances to penetration of chains into the networks and in others to interface reconstruction giving at the interface. To explore the adhesion hysteresis provides by the rubber samples, we have prepared three types of rubber samples: unvulcanized NR, NR with CV and NR with EV. Figure 1 shows the results of the contact mechanics between different rubber samples with glass slide, analyzed with equation 2 to find G as a function of a . We found that all the samples displayed adhesion hysteresis

(different strain energy released rate) between the loading and unloading experiments. However, the amount of that hysteresis increased significantly on going from: NR with EV < NR with CV < unvulcanized NR.

The strain energy release rate (G) measured during the loading experiment (closed symbols) remains independent of a , in good agreement with the expected value of twice the surface energy of control NR (32.0 mJ/m^2). However, the unloading G actually increases as a decrease from the maximum contact radius (corresponding to the maximum load). As the length and flexibility of sulphur crosslinking bridge is increased (CV > EV) the adhesion hysteresis and heat build-up also increase (CV > EV). The vulcanization system of NR samples plays a major role in the adhesion hysteresis and heat build-up. Moreover, unvulcanized NR represents higher adhesion hysteresis and heat build-up than vulcanized NR. This result implies that the mobility of macromolecular chains is an important factor in determining the hysteresis of rubber chain at the interface of substrate and the heat build-up of rubber. The rubber sample with EV should be applied for the energy-saved tyre.

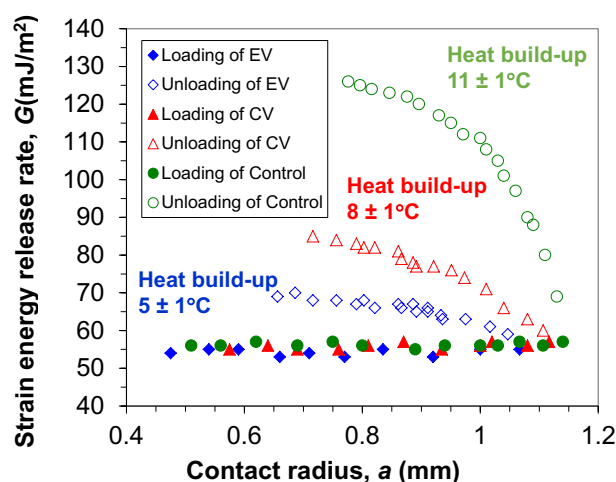


Figure 1. Loading (closed symbols) and unloading (opened symbols) G as a function of contact radius, a , contact with glass slide for three types of rubber samples: unvulcanized NR, NR with CV and NR with EV.

4. Conclusions

The JKR experiment has been well-developed to study the contact mechanics of NR samples with respect of vulcanization system. We found that the adsorption of rubber chains on the surface of glass slide plays a major role in the hysteresis phenomenon. This result indicates a strong dependence of hysteresis and heat build-up on the presence of rubber free chains.

5. References

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