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CHARACTERISATION OF CRACK TOUGHNESS BEHAVIOUR OF UNFILLED AND FILLED ELASTOMERS

W. Grellmann[1], K. Reincke[1], R. Lach[1], G. Heinrich[2]

[1] Martin-Luther-University of Halle-Wittenberg, Department of Engineering Science, Institute of Materials Science, D-06099 Halle, Germany
[2] Continental AG, Strategic Technology Advanced Materials, D-30419 Hannover, Germany

ABSTRACT

With the help of the examinations described in this paper, possibilities should be shown to assess elastomer materials by different fracture mechanics methods. Vulcanizates on the basis of the statistical styrene-butadiene copolymer SBR 1500 with different sulphur and carbon black contents were investigated. Several fracture mechanics examination methods under cyclic, impact-like and quasi-static loading conditions were applied for describing the crack initiation and crack propagation behaviour. The so-called Tear and Fatigue Analyser was used to determine critical values of tearing energy. The instrumented tensile-impact test (ITIT) developed further for elastomer testing is described. By this test, the crack toughness behaviour related to resistance against unstable crack propagation can be examined. At last, a quasi-static fracture mechanics test was applied to the determination of stable crack initiation and crack propagation behaviour. The results of the different tests are discussed in dependence on structure, i.e. sulphur and carbon black content.

KEYWORDS

Fracture Mechanics – Elastomers – Crack Initiation and Propagation – Crack Resistance Curve

INTRODUCTION

Elastomer materials are used for a great number of applications from which defined requirements on material properties follow. Among others, for the application properties of tires for example, even under economic and ecological aspects, wet-skid stability, rolling resistance and wear resistance are important aspects. In the case of passenger car tires, the wear phenomenon mainly appearing is fatigue wear leading to abrasion losses within the tire tread. This is caused by initiation and propagation of cracks from which the application of fracture mechanics concepts for material assessment is derived. The use of fracture mechanics concepts for material characterisation of thermoplastic polymers has been proven to be very helpful in material development and optimization [1]. However, the transfer of these fracture mechanics concepts, such as *J*-integral or COD concept, to elastomeric materials is difficult because of their special, non-linear deformation behaviour. Therefore, fracture mechanics methods have to be modified partly and it is likely necessary to find new ways in analysis of test data recorded. By the experiments described below, it should be shown how usual fracture mechanics test methods work for elastomer testing.

EXAMINATION METHODS

The material behaviour under cyclic loading conditions was tested by using the Tear and Fatigue Analyser (TFA) of Coesfeld GmbH. This is a testing device which was specially developed for the examination of elastomers' fatigue behaviour. Up to ten specimens one-sided cut can be tested at the same time. During the experiment, various measuring parameters are recorded, such as current crack length, load and energy, which serve for subsequent analysis. From these tests, crack propagation curves for each material were plotted and critical tearing energies as a measure for the materials' resistance against fatigue crack propagation were determined. Instrumented tensile-impact tests (ITIT) can be used to investigate even such flexible materials like elastomers under impact-like loading conditions. In principle, a specimen is fixed between the unsecured crosshead and the secured clamp (see Figure 1). Then the specimen is loaded by the pendulum hammer that impacts the unsecured crosshead, and so the specimen is strained in direction of its longitudinal axis until it tears. At the same time the load–time curve is recorded, and afterwards through double integration the load-extension curve is calculated. In analogy to the instrumented *Charpy* impact test (ICIT) [2], characteristic parameters are used for analysis, and J values J_d were determined according to an evaluation method of Begley and Landes.

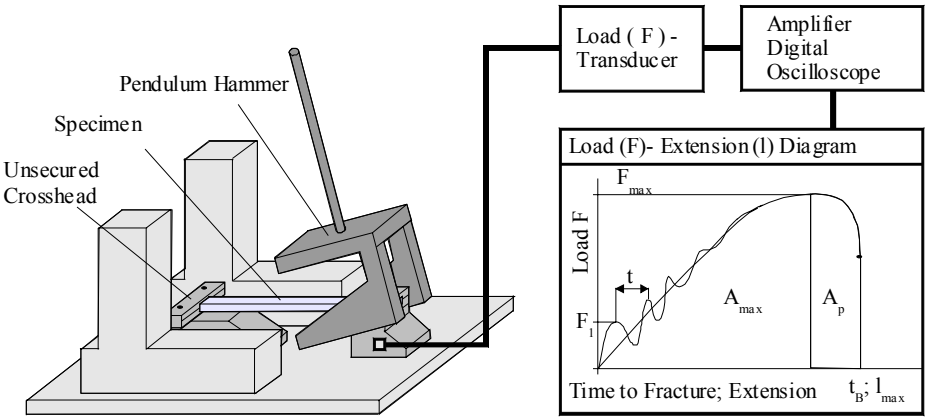

Figure 1: Schematic representation of the instrumented tensile-impact test

Besides these two methods described above, recording of crack resistance (R-) curves using a quasi-static fracture mechanics test took place. Following the stop-block method of the ICIT, the tests on several specimens (multiple-specimen method) were stopped after reaching different strain values to produce different amounts of stable crack growth. The fracture surfaces were investigated with a light microscope to determine the size of stable crack growth. From the recorded load–extension curves, energies for calculation of the loading parameter J for the R-curve were determined. Finally, the J–Δa data were plotted and regression functions were fitted and then used for subsequent analysis. Ascertainable parameters of the crack resistance curves are, for example, technical crack initiation values $J_{0.2}$ determined according to ESIS TC 4 recommendation [3] or the slope of the R-curve $dJ/d\Delta a$. These parameters supply quantitative criteria for a material comparison.

MATERIALS

Basis of the materials investigated is the statistical styrene-butadiene copolymer SBR 1500 with a styrene content of about 23 wt.-%. Crosslinking occurred with a sulphur–accelerator system. The sulphur content was varied in the range from 0.8 to 2.4 parts per hundred rubber (phr) with a constant sulphur–accelerator ratio to examine the influence of the crosslink density on the properties. For assessment of filler influence, vulcanizates with a constant sulphur content of 1.6 phr, but with different carbon black contents in the range from 0 to 50 phr were produced. Vulcanizates of this kind are used in different areas, for example in tires or conveyor belt materials. For these materials, network densities (see Figure 2) were determined on the basis of the Mooney-Rivlin equation using stress–strain diagrams. The physical network density shown in Figure 2 contains a primary network density as a result of chemical crosslinking and elastically active entanglements. In

the case of filled vulcanizates, a secondary apparent network density of the filler network can be estimated that is related to elastically effective filler-polymer interactions and to the contribution of the filler–filler networking above a certain percolation threshold.

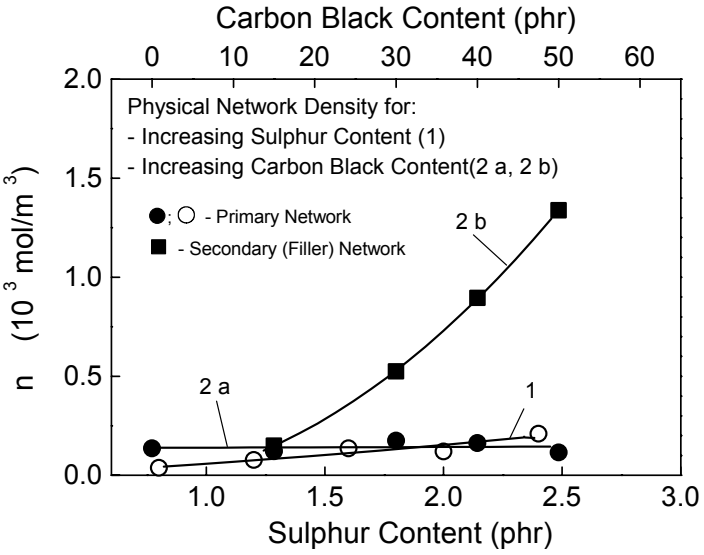

Figure 2: Network density of the elastomers examined

For the fatigue tests, SENT specimens were used with the dimensions of length l = 64 mm, width W = 15 mm and thickness B = 1.5 mm. The initial crack length was about 1 mm. For the tensile-impact tests and for the quasi-static fracture mechanics tests, DENT specimens were used which had the same dimension as for the fatigue tests, but the initial crack length was about 4 mm and 6 mm, respectively.

RESULTS

Critical tearing energy values T_c determined by the TFA measurements and the results of the instrumented tensile-impact test for the unfilled vulcanizates are shown in Fig. 3. Because T_c remains nearly constant up to a sulphur content of 2.0 phr, it can be said that the increasing network density (see Fig. 2) in this sulphur content range has no influence on T_c. Only a higher sulphur content decreases the critical tearing energy. The J values indicate a constant decrease of toughness, e.g. the resistance against unstable crack propagation is decreased with increasing network density.

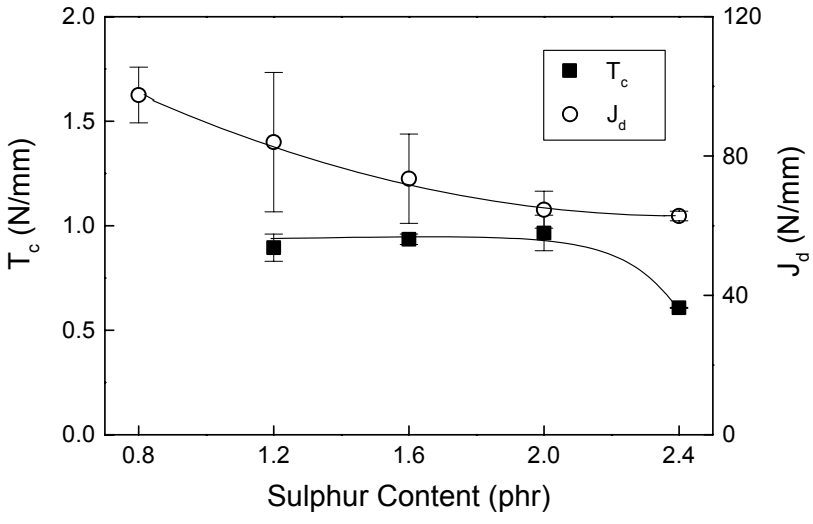

Figure 3: Results of the cyclic and impact tests for the unfilled vulcanizates

However, the T_c values and the J values of the filled vulcanizates are decisively influenced by changes in structure, i.e. by the rising carbon black content. This is attributed to reinforcing effects and the development of a filler network and increasing interactions between filler and polymer as well as filler and filler, respectively. For the J values of the filled materials a maximum was found at 40 phr carbon black. From the maximum loads and maximum extensions is derived that this maximum is strength-determined and shows that only a filler content up to 40 phr leads to an improvement of the crack growth behaviour of the observed material system. Possible causes for the decreasing parameter level with 50 phr carbon black are, on the one hand, reduction of effective network chain length due to the high filler content and a reduced extensibility in the area of stress concentrations within the material, which decrease the energy absorption capacity. On the other hand, an incomplete distribution of the carbon black particles and agglomerates, respectively or larger filler agglomerates are considered as inhomogeneities and work as crack starter due to unfavourable stress circumstances nearby.

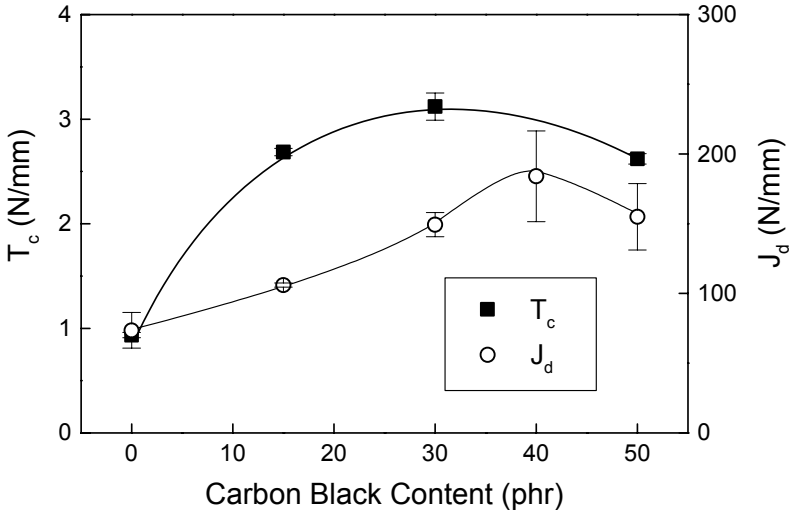

Figure 4: Influence of carbon black content on T_c and on J_d values related to resistance against unstable crack propagation

The results of the quasi-static fracture mechanics examination are shown in Figs. 5 and 6. Figure 5 represents the J–Δa curves, where a significant increase of the slope with rising carbon black content is visible. In comparison with the filled materials, only small differences resulted for the unfilled vulcanizates. This is reflected by the technical crack initiation values $J_{0.2}$ and the values of the slope of the crack resistance curves $dJ/d(\Delta a)$ at the point of maximum experimentally determined crack growth $\Delta a_{\max\,(exp)}$ (Figure 6).

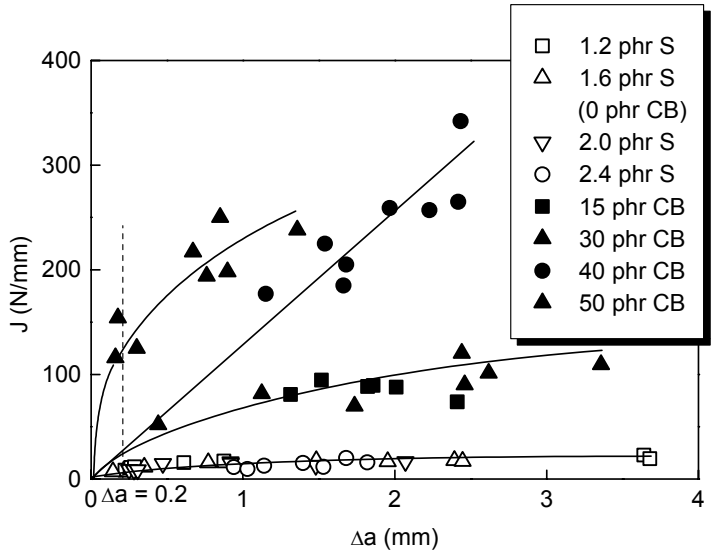

Figure 5: Crack resistance curves

The crack initiation and crack propagation behaviour is only slightly influenced up to a sulphur content of 2.0 phr, a still higher sulphur content of 2.4 phr leads to a lower crack initiation value, but to a higher resistance against crack propagation (see Figure 6a).
With an increasing carbon black content up to 30 phr, $J_{0.2}$ is slightly increased at first, then a strong increase of the resistance against stable crack initiation appears. Even the resistance against stable crack propagation characterised by $dJ/d(\Delta a)$ at $\Delta a_{max(exp)}$ rises slightly up to 30 phr filler and shows a maximum value at 40 phr. That means, more energy is necessary to initiate a stable crack within the 50 phr filled material, but in comparison with the 40 phr filled vulcanizate, the resistance against stable crack propagation is lower.
The observed differences of the crack initiation and propagation behaviour show that a multi-parametrical description of the fracture behaviour is necessary for an optimal material characterisation.

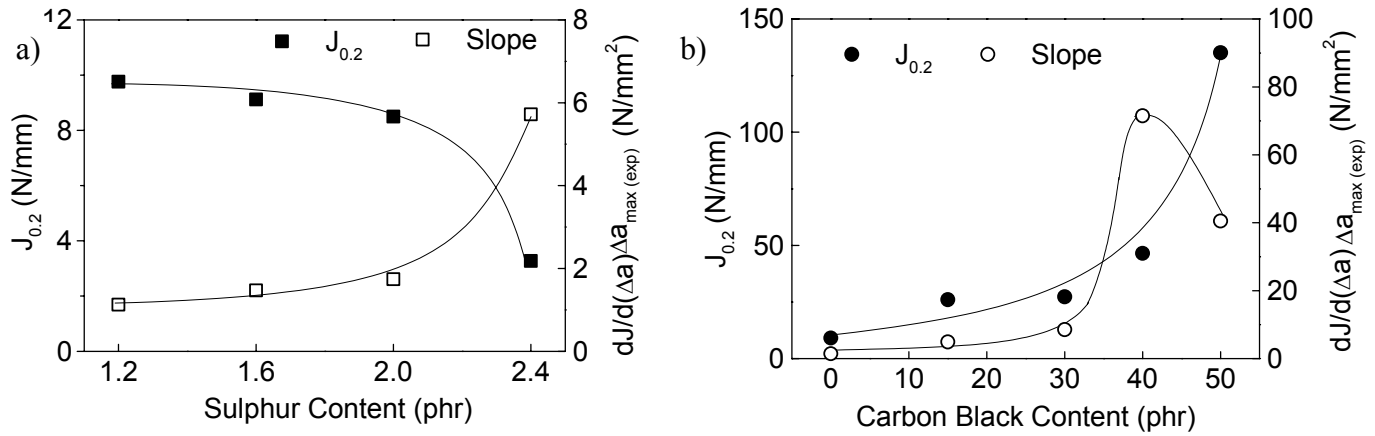

Figure 6 a, b: Influence of sulphur **(a)** and carbon black content **(b)** on the technical crack initiation value and the slope of the R-curves at the point $\Delta a_{max\ (exp)}$

CONCLUSIONS

With the examination methods described here, one can assess the toughness behaviour of the elastomer materials investigated. An increase of the sulphur content, this means an increase of crosslink density, leads, in some cases, to a considerable decrease of the toughness parameters determined under various loading conditions. In dependence on filler content, maximum values for the various toughness parameters, except for the technical crack initiation parameter $J_{0.2}$, were found at 30 and 40 phr filler content, respectively. Therefore, the addition of 30–40 phr carbon black shows in our study the optimal improvement of the crack initiation and propagation behaviour of this elastomer system in the filler content range investigated.

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