



# Crack-tip shape in the crack-growth rate transition of filled elastomers



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## ABSTRACT

The velocity transition in crack-growth dynamics and the accompanying changes in crack tip shapes are investigated for the elastomers composed of styrene-butadiene rubber and silica filler (SBR/SI). The concentrations of filler and cross-link ( $\phi_f$  and  $c_x$ , respectively), and temperature ( $T$ ) are extensively varied in order to change the degree of nonlinearity in elasticity of the elastomers. The shapes of the crack tip are characterized by three parameters, i.e., the deviation  $\delta$  from the parabolic one expected by the linear elastic fracture mechanics (LEFM), the parabolic curvature  $a$ , and the non-dimensional quantity  $a\delta$ . The dependence of these parameters on the input tearing energy  $I$  is successfully explained by the weakly nonlinear theory of dynamic fracture (WNLT), which considers the contribution of the second-order shear modulus  $\mu_{(2)}$  to the strain field in addition to that of the first-order shear modulus  $\mu_{(1)}$ , independently of  $\phi_f$ ,  $c_x$  and  $T$ , unless  $\delta$  exceeds a limit value ( $\delta_C$ ). SBR/SI shows an appreciably larger value of  $\delta_C$  than the carbon-black filled acrylonitrile butadiene (NBR/CB) elastomers. The smaller value of  $\delta_C$  for NBR/CB is attributed to that the contribution of the third-order nonlinear modulus  $\mu_{(3)}/\mu_{(1)}$ , which is not considered in WNLT, is appreciably higher than that in SBR/SI. The magnitude of the threshold tearing energy  $I_c$  for the onset of the velocity transition shows a good correlation with the fracture toughness  $W_c$  normalized by the nonlinear elastic constant  $\mu_{(1)}^3/\mu_{(2)}^2$  in WNLT. This result indicates that the magnitude is governed by the combined effect of the degree of nonlinearity in elasticity and the fracture toughness.

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## 1. Introduction

The failure of rubber materials often occurs via the propagation of the cracks which originate from defects. Understanding of the crack-growth dynamics is particularly important in practical and industrial applications of rubber, because the suppression of the initiation as well as the subsequent propagation of cracks are crucially required to prolong the life of industrial rubber products such as tire.

Although the crack-growth in elastomers has been studied for many years, many fundamental problems are still left unsolved. An important problem is how mechanical properties of elastomers such as elasticity, viscoelasticity, and their nonlinearity correlate with the features of crack-growth phenomena such as propagating velocity and shapes of crack tip. It has been realized that the

nonlinearity of elasticity and viscoelasticity has finite effects on crack-growth dynamics, but the details of these effects remain to be fully characterized [1–10].

One of the spectacular aspects in crack-growth phenomena of elastomers is an abrupt change in the crack-growth rate more than two orders of magnitude in a very narrow range of the input tearing energy [11–13]: The propagating velocity ( $v$ ) undergoes a discontinuous-like transition between the “slow mode” (typically,  $v \approx 10^{-5} - 10^{-3}$  m/sec) and the “fast mode” (typically,  $v \approx 10^{-1} - 10^2$  m/sec). This velocity transition phenomenon has attracted much interest from the fundamental and practical viewpoints, and it has been investigated both experimentally [11–15] and theoretically [16–18] for more than 30 years as a classical issue in the physics of rubber. The earlier studies have revealed some complicated aspects of the velocity transition phenomenon. For instance, the threshold tearing energy for the onset of the transition depends on temperature, but the temperature dependence of the threshold value cannot be explained by the familiar time-temperature superposition principle using the linear viscoelasticity of the bulk elastomers [12,13]. This result indicates that the velocity transition is not associated simply with the linear viscoelasticity of the bulk

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elastomers.

The shapes of crack tip involve important information about the mechanism of crack propagation, because they directly reflect local strain/stress field near the tip. It has been known that the observed crack tip shapes often significantly deviate from a parabolic one which is expected by linear elastic fracture mechanics (LEFM) [15,19–21]. The observed power-law relation between the tearing energy and crack-growth rate [12,22] has been successfully explained by a theory which introduces the linear viscoelastic effect into LEFM [23–26], but the non-parabolic shapes of the crack tip cannot be described by LEFM and the LEFM-based theories. Recently, Bouchbinder, Livne and Fineberg et al. proposed the weakly nonlinear theory of dynamic fracture (WNLT) which considered the second-order elastic contribution to the strain field around the crack tip in addition to the first-order one [27–29]. They showed that WNLT explained the features of the non-parabolic shapes of the crack tip observed for non-dissipative polymer gels [27–29]. Their results on polymer gels imply that the nonlinear elasticity has also an important role in the crack-growth dynamics of elastomers with finite dissipation.

In the previous study [30], we have revealed for the carbon-black-filled elastomers that the velocity transition is accompanied by a significant change in the shapes of crack tip. Our home-built experimental setup enabled us to measure simultaneously the crack-growth rate and the crack tip shape as a function of the input tearing energy, which type of tests is usually referred to as “ $dc/dt$ ”, and in this test, a relatively large deformation is applied to a sample, and the crack grows continuously with time at a rate of  $dc/dt$ , where  $c$  is the crack length and  $t$  is time [12,19,22,31–39]. The crack tip shapes pronouncedly changed from blunt to sharp shapes in relation to the velocity transition from the slow mode to the fast mode [30]. Excepting the slow mode region of the elastomers with very low carbon-black contents, the crack tip shapes appreciably deviated from the parabolic one, and the deviation became larger with an increase in velocity. The crack tip shapes were characterized by two specific parameters, the deviation  $\delta$  from the parabolic shape and the parabolic curvature  $a$ . The dependence of  $\delta$  and  $a$  on tearing energy was successfully explained by WNLT [27–29] when  $\delta$  was not greater than 1 mm, independently of the crack-growth mode (slow or fast) [30]. In this analysis, the ratio of the first-order and second-order shear moduli ( $\mu_{(2)}/\mu_{(1)}$ ), which was evaluated from the nonlinear stress-strain relations, was employed as a measure of the degree of nonlinearity in elasticity for the bulk materials. Our previous work demonstrated the importance of the nonlinear elasticity of bulk elastomers in the velocity transition phenomena.

These results in the previous study [30] raise several further issues. One is the effects of the environmental and material parameters which influence the degree of nonlinearity in elasticity. Each of temperature, the concentrations of cross-link and filler, and the kinds of rubber and filler significantly affects the nonlinear stress-strain behavior of elastomers. It has been reported that the major features of the velocity transition including the threshold tearing energy are influenced by some of these variables [11,12,15]. In the previous study [30], the filler content was varied, but the cross-linker concentration was fixed and all measurements were conducted at a fixed temperature for only one type of elastomer composed of acrylonitrile butadiene copolymer rubber (NBR) and carbon-black (CB) filler. The second issue is the physical origin of the upper limit value of  $\delta$  ( $\delta_c$ ) for the applicability of WNLT in the interpretation of the crack tip shapes. The previous work [30] evaluated  $\delta_c$  to be about 1 mm, while the thickness of the specimens was 1 mm. It has a possibility that  $\delta_c$  might be affected by the specimen thickness. The investigations for the thickness effects on the velocity transition, especially on the crack tip profiles are required to clarify this issue. The third is the governing factors for

the magnitude of threshold tearing energy for the velocity transition ( $T_c$ ). This transition phenomenon has long been investigated, but it still remains to be understood what kind of properties of bulk elastomers governs the magnitude of  $T_c$ . This issue is also practically important for the life prolongation design of rubber products.

The present work investigates the velocity transition in crack-growth dynamics and the accompanying changes in crack tip shapes for the silica-filled styrene-butadiene rubber (SBR/SI elastomers) which are different in chemical structures of rubber and filler from the NBR/CB elastomers in the previous work [30]. The concentrations of filler and cross-link are widely varied, and temperature is also extensively altered from a rather low temperature of  $-20$  °C to a high temperature of 100 °C. The specimen thickness is also varied from 0.7 mm to 2.0 mm. The data for the specimens with varying systematically the values for several parameters provide a definite basis to discuss the three issues described above. The results in present study will contribute to the deep understanding of the physics of crack-growth dynamics in elastomers, and they will also give the useful and fundamental information for the life prolongation design of rubber products.

## 2. Experiment

### 2.1. Samples

The solution styrene butadiene copolymer (styrene content is 25%, TUFENE 2000, Asahi Kasei Corporation) and silica (Nipsil, Tosoh Silica Corporation) were employed as rubber matrix and filler, respectively. Bis-(triethoxysilylpropyl)-polysulfide (Shin-Etsu Chemical Corporation) was used as silane coupling agent. The filler (silica) contents ( $\phi_f$ ) were varied via the volume fraction in the polymer matrix from 0.05 to 0.21. The concentrations of sulfur for cross-linking ( $c_x$ ) were varied from 1.05 wt% to 1.75 wt% to polymer matrix. The silica-filler content was fixed to 0.14 for the investigation on the effects of cross-link densities, while the sulfur concentration was fixed to 1.4 wt% for the investigation on the effects of filler content.

Sheet samples were used for tensile tests and crack-growth measurements, and the dimensions of sheet samples were ( $x \times y \times z$ )  $180 \times 20 \times 1$  mm, where  $x$  was the crack-growth direction,  $y$  was the loading direction, and  $z$  was the specimen thickness direction, respectively (Fig. 1). The specimen thickness was 1 mm unless specified otherwise whereas the thickness was varied from 0.7 to 2.0 mm.

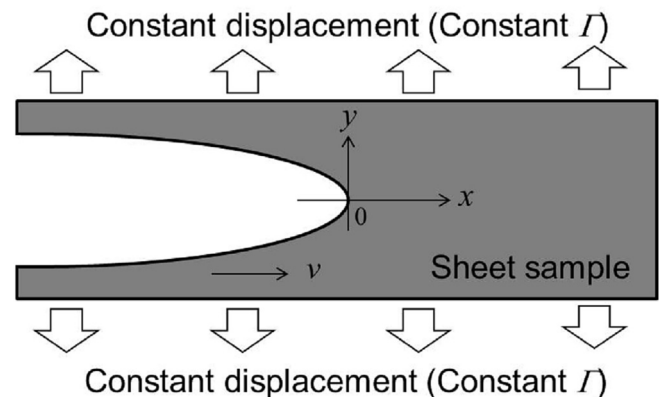


Fig. 1. Experimental geometry for the crack-growth dynamics: the  $x$  axis is the crack growth direction and the  $y$  axis is the loading direction.

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