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Flaw sensitivity of highly stretchable materials

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A B S T R A C T

Elastomers and gels can often deform multiple times their original length. The stretchability is insensitive to small cuts in the samples, but reduces markedly when the cuts are large. We show that this transition occurs when the depth of cut exceeds a material-specific length, defined by the ratio of the fracture energy measured in the large-cut limit and the work to rupture measured in the small-cut limit. This conclusion generalizes a result in the fracture mechanics of hard materials. For an acrylic elastomer and a polyurethane, we measure the stretch to rupture as a function of the depth of cut, and show that the experimental data agree well with the prediction of the nonlinear elastic fracture mechanics. In a space of material properties we compare many materials (elastomers, gels, ceramics, glassy polymers, biomaterials, and metals), and find that the material-specific length varies from nanometers to centimeters.

1. Introduction

Stretchable materials, such as elastomers and gels, have long been used in tires, seals, gloves, and contact lenses. Under development are new fields of applications, including tissue regeneration [1], drug delivery [2], artificial muscles [3–5], stretchable electronics [5–9], and soft robots [10,11]. Stretchable, transparent, ionic conductors (e.g., hydrogels and ionogels) enable devices of unusual functions, such as transparent loudspeakers [12], artificial skins [13], artificial axons [14,15], and electroluminescence of giant stretchability [16–18]. The interest in the mechanics of stretchable materials has surged [19–32].

This paper focuses on a specific issue in the mechanics of stretchable materials: the reduction of stretchability by cuts. A cut can be introduced either intentionally using a razor blade, or unintentionally during fabrication. In the latter case, the cut is commonly called a *flaw*. The reduction of stretchability by cuts and flaws is called *flaw sensitivity*. For example, an acrylic elastomer, VHB™, commonly used in the development of artificial muscles [3], can deform beyond ten times its original length [33]; however, a VHB sample containing a cut of a few millimeters ruptures when deforming 3–5 times its original length [34]. As another example, a recent tough hydrogel can deform more than twenty times its original length, and a centimeter-long cut reduces the stretchability to seventeen times [26].

Two approaches exist to predict the rupture of a stretchable device. In one approach, the designer assumes a flawless device, calculates the field of deformation using the nonlinear theory of elasticity, and predicts rupture if any material point in the device reaches a critical state of deformation [35–43]. In the other approach, the designer identifies a flaw in the device, calculates the energy release rate using the nonlinear theory of elasticity, and predicts rupture if the energy release rate reaches the fracture energy [44–47].

The two approaches work well in two limits. The first approach requires no knowledge of flaws, and is applicable in the limit of small flaws. The second approach requires the knowledge of flaws, and is applicable in the limit of large flaws. The transition from flaw-insensitive to flaw-sensitive rupture has been discussed in the literature [48–50], but the size of the flaws over which the transition occurs is vague for stretchable materials.

Here we study the transition from flaw-insensitive rupture to flaw-sensitive rupture of highly stretchable materials. For an uncut sample, we measure the work to rupture, W_*, which has the dimension of energy per unit volume. For a sample with a large cut, we measure the fracture energy, Γ, which has the dimension of energy per unit area. The ratio of these two parameters, Γ/W_*, defines a material-specific length, which we call the *length of flaw sensitivity*. Using a combination of experiment and calculation, we show that this material length marks the transition from flaw-insensitive to flaw-sensitive rupture. When the depth of cut c

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is small compared to Γ/W_*, the stretchability is insensitive to the cut. When c is large compared to Γ/W_*, the stretchability reduces markedly as the depth of the cut increases. Furthermore, we show that flaw sensitivity depends on the stretch-stiffening behavior of elastomers, and that the experimental data agree well with the prediction of nonlinear elastic fracture mechanics. The concept of flaw sensitivity is applicable to all materials, including metals, ceramics, biomaterials, and polymers. We represent the lengths of flaw sensitivity of various materials in a space of material properties, with W_* and Γ as axes.

The length Γ/W_* has been used to characterize the intrinsic diameter of the crack tip in elastomers [51,52], but has not been used to characterize flaw sensitivity. We next compare Γ/W_* to other material lengths commonly used in fracture mechanics. A length, $\Gamma/\left(\sigma_*^2/E\right)$, appears in the crack-bridging model, where σ_* is the maximum stress in the traction–separation curve [53–58]. In the crack-bridging model, the region outside bridging zone is linearly elastic with Young's modulus E. The work to rupture near the crack tip is given by the $W_* = \sigma_*^2/2E$. For highly stretchable materials, however, the material outside the bridging zone is nonlinearly elastic. The material length Γ/W_* generalizes $\Gamma/\left(\sigma_*^2/E\right)$ when linear elasticity does not apply. (We have dropped any numerical factor.) Another frequently discussed material length is the ratio of fracture energy and elastic modulus, Γ/E [32,59]. This length overestimates the length of flaw sensitivity by orders of magnitude. For a highly stretchable material, the stretchability λ_* is on the order of ten, so that $W_* \gg E$. Consequently, the length of flaw sensitivity Γ/W_* is much smaller than Γ/E.

2. Transition from flaw-insensitive to flaw-sensitive rupture

Flaw-insensitive rupture

To focus on essentials, we consider the stretchability of a thin sheet of a material under a uniaxial force. The length and the width of the sheet are much larger than the thickness of the sheet and the depth of the cut. Using an uncut sample, we measure the applied force as a function of the associated displacement. The area under the force–displacement curve divided by the volume of the material defines the energy density, W. Let λ be the stretch, namely, the length of the deformed sheet (in the direction of the applied force) divided by the length before stretch. The energy density is a function of stretch, $W(\lambda)$.

For an uncut sample, let λ_* be the stretch to rupture and W_* be the work to rupture. The two parameters are related by the function $W(\lambda)$:

$$W_* = W(\lambda_*). \tag{1}$$

The stress to rupture σ_* is defined by the applied force at rupture divided by the deformed cross-sectional area (perpendicular to the applied force). Criterion (1) also applies to samples containing cuts small compared to a material length (to be specified).

The stretchability of elastomers is insensitive to small flaws. Table 1 summarizes the experimental data from the literature and from this work. The reported stretch, stress, and work to rupture are within variations 5%–20% of their means. These data were measured using uncut samples. Yet flaws exist inevitably, either as small cracks or as heterogeneities of materials [50]. The small scatter in the data indicates that the stretchability of these materials is insensitive to the small flaws. This observation on elastomers differs from that on brittle hard materials, e.g., silica glass, in which a micron-sized flaw reduces the strength by orders of magnitude [44].

Flaw-sensitive rupture

Flaw-sensitive rupture is predicted by fracture mechanics [47]. Consider a sheet containing a cut. The elastic energy of the sample is a function $U(\Delta, c)$, where c is the depth of the cut in the undeformed state, and Δ is the displacement associated with the applied force. The reduction in the elastic energy when the cut extends a unit area defines the energy release rate, $G = -\partial U(\Delta, c)/(t\partial c)$, where t is the thickness of the sheet in the undeformed state. The energy release rate can be determined by solving the boundary-value problem of nonlinear elasticity. When other sizes of the specimen are much larger than the cut, the depth of cut c is the only length scale in the boundary-value problem. Dimensional considerations dictate that the energy release rate should take the form, $G(\lambda, c) = k(\lambda) W(\lambda) c$, where $k(\lambda)$ is a dimensionless function determined by solving the boundary-value problem. The function $k(\lambda)$ depends on the model of nonlinear elasticity [47,60]. The sample ruptures at stretch λ_R when the energy release rate reaches the fracture energy, $G(\lambda_R, c) = \Gamma$. When the cut is large and the sample ruptures at a small applied strain, $\lambda \to 1$, linear elasticity applies, and the small-deformation limit for an edge cut is known, $k(1) \approx 2(1.1215)^2 \pi \approx 7.9$ [61]. The criterion of rupture becomes

$$W(\lambda_R) = \frac{\Gamma}{k(1)\, c}. \tag{2}$$

This result is the Griffith limit [44]. Thus, we characterize the flaw-sensitive rupture by the fracture energy Γ in the limit of large flaws.

The transition from flaw-insensitive rupture to flaw-sensitive rupture

We have characterized the rupture of an uncut sample by the work to rupture, W_*, which has the dimension of energy per unit volume. For a sample containing a cut, the state W_* prevails ahead the front of the cut when the sample is near rupture. We have also characterized the rupture in the limit of large cuts by the fracture energy, Γ, which has the dimension of energy per unit area. The ratio of these two material parameters defines a material-specific length, Γ/W_*.

We argue that the material length Γ/W_* marks the transition from flaw-insensitive to flaw-sensitive rupture. When a sample of a small cut ruptures, except for the unstrained region behind the front of the cut, the entire sample reaches the state of W_* (Fig. 1(a)). When a sample of a large cut ruptures, only a small zone around the front of the cut reaches the state of W_* (Fig. 1(b)). Inside this zone, fracture process occurs. Outside this zone, the field of deformation is well characterized by the nonlinear theory of elasticity. The length scale of the fracture process zone is estimated as follows. A dimensional consideration dictates that energy density W should scale with the energy release rate G, and inversely scale with the distance to crack tip r, namely, $W \sim G/r$. This scaling appears in the analytical solutions of the nonlinear elastic field around the front of cut [25]. When the sample ruptures, the energy release rate G reaches Γ, and the energy density W in the fracture process zone attains W_*. Consequently, the size of the fracture process zone scales with the material length Γ/W_*.

A flaw-sensitivity diagram displays the stretch to rupture λ_R as a function of the depth of cut c (Fig. 1(c)). The transition occurs when the depth of cut c is comparable to the material length Γ/W_*. When the depth of the cut c is small compared to Γ/W_*, the stretch to rupture is insensitive of the depth of the cut, and the small-cut limit (1) applies. When the depth of cut is large compared to the material length, the stretch to rupture decreases as the depth of the cut increases, and approaches the large-cut limit (2).

The material length Γ/W_*, together with the depth of the cut c, defines a dimensionless number:

$$\chi = \frac{c}{\Gamma/W_*}, \tag{3}$$