



# Smoothing creases on surfaces of strain-stiffening materials



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## ABSTRACT

When an elastic block (e.g., an elastomer or a soft tissue) is compressed to a critical strain, the smooth surface of the block forms creases, namely, localized regions of self-contact. Here we show how this instability behaves if the solid stiffens steeply. For a solid that stiffens steeply at large strains, as the compression increases, the surface is initially smooth, then forms creases, and finally becomes smooth again. For a solid that stiffens steeply at small strains, creases will never form and the surface remains smooth for all levels of compression. We also obtain the critical conditions for the onset of wrinkles. When the surface does become unstable, we find that creases always set in at a lower compression than wrinkles. Our findings may shed light in developing crease-resistant materials

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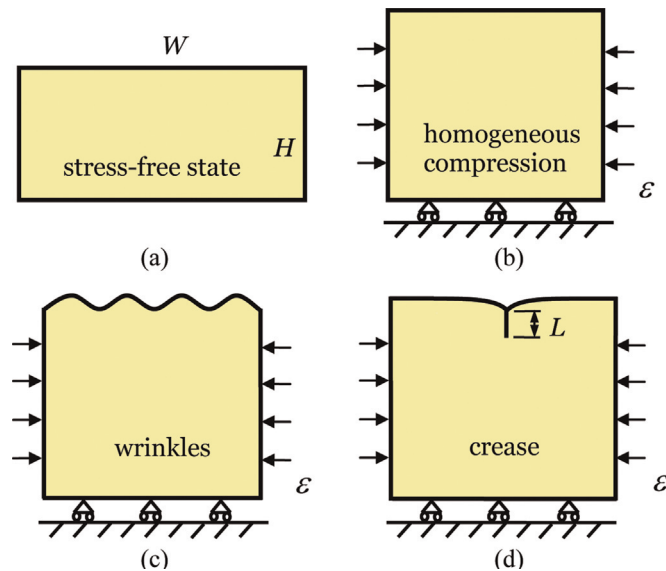
## 1. Introduction

Biot (1963) analyzed an elastic block compressed under the plane strain conditions (Fig. 1a and b), and predicted that the flat surface of the block was unstable when the compression reached a critical strain of 0.46. This theoretical prediction remained unchallenged until Gent and Cho (1999) noted its disagreement with their experimental finding that the surface formed creases at a critical strain of 0.35. Hohlfeld (2008) and Hohlfeld and Mahadevan (2011) showed that Biot's solution and creases are two distinct instabilities, and that creases set in at a critical strain of 0.35. Biot linearized the boundary-value problem around a state of finite homogeneous deformation, and his solution corresponds to a smooth, wavy surface (i.e., wrinkles) of small strain relative to the homogeneous state (Fig. 1c). By contrast, a crease is a localized, self-contact region of large strain relative to the homogeneous state (Fig. 1d). The critical strain for the onset of creases has since been obtained by several other approaches of numerical analysis (Hong et al., 2009; Wong et al., 2010; Cai et al., 2010; Hong and Gao, 2013; Tallinen et al., 2013). Furthermore, Hohlfeld (2013) mapped the onset of a crease to the coexistence of two scale-invariant states. A post-bifurcation analysis of Cao and Hutchinson (2012) showed that Biot's solution is unstable.

No evidence exists that Biot's smoothly wavy surfaces have ever been observed experimentally on homogeneous elastic blocks under compression. Creases, however, have been observed routinely on elastic blocks compressed by various means, including mechanical forces (Cai et al., 2012; Gent and Cho, 1999; Ghatak and Das, 2007; Mora et al., 2011), constrained swelling (Arifuzzaman et al., 2012; Barros et al., 2012; Dervaux and Ben Amar, 2012; Dervaux et al., 2011; Guvendiren et al., 2010; Ortiz et al., 2010; Pandey and Holmes, 2013; Tanaka, 1986; Tanaka et al., 1987; Trujillo et al., 2008; Weiss et al., 2013; Zalachas et al., 2013), temperature change (Kim et al., 2010), electric fields (Park et al., 2013; Wang et al., 2012; Wang et al., 2011; Wang and Zhao, 2013a; Xu and Hayward, 2013), and light (Yoon et al., 2012). Creases have been studied in soft tissues

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**Fig. 1.** (a) In the stress-free state, a block of elastic material is of width  $W$  and height  $H$ . Under compression the material may deform in several ways. (b) The block undergoes homogeneous compression. (c) The surface forms wrinkles. (d) The surface forms a crease of depth  $L$ . The applied strain  $\epsilon$  is defined by the compressive displacement divided by the initial width of the block.

(Bayly et al., 2014; Jin et al., 2011; Yang et al., 2007). Creases have been related to the Schallamach waves arising during the frictional sliding of a rubber against a rigid surface (Gabriel et al., 2010), and to the osmotic collapse of a water-filled cavity in a hydrogel (Cai et al., 2010). Creases can also form on the interface between two elastic solids (Jin et al., 2014). Applications of creases have been explored, including the use of creases to control chemical patterns (Kim et al., 2010; Yoon et al., 2012), enzymatic activity (Kim et al., 2010), cellular behavior (Saha et al., 2010), adhesion (Chan et al., 2011), and biofouling (Shivapooja et al., 2013).

Although wrinkles have never been observed experimentally on large homogeneous elastic blocks under compression, many factors affect the behavior of creases, and may even promote the formation of wrinkles. Surface energy adds a barrier to the nucleation of creases, and makes nucleation defect-sensitive (Chen et al., 2012; Yoon et al., 2010). When the loading is an electric field, wrinkles may form when the elastocapillary effect is strong enough (Wang and Zhao, 2013a). For a layer of finite thickness with a traction-free bottom surface, creases on the top surface are subcritical—that is, as the applied compressive strain increases and then decreases, creases form and disappear with hysteresis (Hohlfeld and Mahadevan, 2012). For a stiff film on a soft substrate under compression, the film forms periodic wrinkles at a small strain (Bowden et al., 1998). As the strain increases, the wrinkles double their period, and ultimately lead to deep folds (Pocivavsek et al., 2008). When the film and the substrate have comparable moduli, the transitions between creases, wrinkles and folds become complex (Hutchinson, 2013; Wang and Zhao, 2013b). Complex behavior also occurs in a solid of gradient modulus (Diab et al., 2013; Wu et al., 2013). If the substrate is pre-compressed, creases are subcritical, and form and disappear with hysteresis (Chen et al., 2014).

Biot's original analysis, as well as much of the subsequent theoretical work, represents the elastic solid by the neo-Hookean model. This model describes elastomers of long polymer chains well, but is inadequate when materials stiffen steeply. A soft biological tissue, for example, is usually a composite of a compliant matrix and stiff fibers (Fung, 1993). When the tissue is under a small strain, the matrix carries much of the load, but the fibers are not tight, so that the tissue is soft. As the strain increases, the fibers gradually tighten and rotate to the loading direction, so that the tissue stiffens steeply. As another example, an elastomer is a three-dimensional network of long and flexible polymer chains (Treloar, 1975). When the elastomer is under no stress, the chains undergo thermal motion and coil. When the elastomer is subject to moderate strains, the chains uncoil and the stress-strain relation of the elastomer is well represented by the neo-Hookean model, which is derived under the assumption of Gaussian chains. When the chains become nearly straight, however, they no longer obey the Gaussian statistics, and the stress-strain curve rises steeply and deviates significantly from the neo-Hookean model. Destrade et al. (2009) analyzed the onset of wrinkles on the surface of a bending block. They showed that when the material stiffens at a relatively small strain, the critical strain for the onset of wrinkles differs significantly from that of a neo-Hookean material. These authors, however, did not consider the formation of creases.

Here we represent a strain-stiffening material by using the Gent model (Gent, 1996), and study the initiation and development of creases by using a finite element method. For a solid that stiffens at large strains, as the compression increases, the surface is initially smooth, then forms creases, and finally becomes smooth again. For a solid that stiffens at small strains, creases will never form and the surface remains smooth for all levels of compression. We also study the condition for the

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