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Effect of far-field compliance on local failure dynamics of soft solids

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ABSTRACT

The resistive forces and failure of a soft gel upon large strain indentation with small, spherically-tipped indenters (or puncture mechanics) has been studied previously by Fakhouri et al., (2015). The above work is extended to include the effect of far-field compliance on the critical crack initiation event via puncture in soft acrylic triblock copolymer gels. It is found that critical puncture force, P_c, reduces with increasing far-field compliance, C_s, due to the change in local probe velocity, v_g. A power-law relation between P_c and C_s is established and is shown to be valid across a range of global probe velocity, v_c, varied over three orders of magnitude. Small-strain mechanical response of the gels characterized via oscillatory shear rheology and large strain response measured from puncture mechanics show rate-independent behavior. Our experiments and analysis provide insight into local rate-dependent failure processes in elastic triblock copolymer gels.

1. Introduction

Soft polymer networks in the form of elastomers, gels, and synthetic mimics of biological tissues have become increasingly prevalent for applications in medicine, food science, electronics, and adhesion [1–5]. The growing demand for soft polymer gels and elastomers increases the importance of developing an enhanced understanding of their mechanical behavior. Classical theories such as Linear Elastic Fracture Mechanics (LEFM) have been well established to study fracture in conventional hard materials [6]. Soft polymer gels, on the other hand, undergo large deformations prior to failure, which leads to elastic crack blunting at the crack tip [7]. Therefore, understanding failure in soft materials with LEFM is challenging because it requires knowledge of large strain mechanical response of these materials [5,8–10]. Fracture mechanisms in soft solids are also known to be sensitive to several characteristic length scales governed by material parameters at which forces due to surface tension and adhesion can possibly resist crack growth in addition to elasticity [11–13]. For example, two such length scales are the elasto-capillary length, Υ/E (determined by the surface stress, Υ, and elastic modulus, E) [12,14–16], and the elasto-fracture length, G_c/E given by the ratio of fracture toughness (or critical energy release rate for crack propagation), G_c and E [11]. Moreover, fracture in polymer gels and elastomers is known to be rate dependent mainly due to viscoelastic dissipative processes in the bulk [17–19], rate dependent crack tip processes such as viscous chain pull-out [20,21], poroelastic relaxations corresponding to migration of the solvent within the network [22–24], and kinetics of bond rupture near the crack tip region [25–27].

Although the above stated challenges have been addressed to a large extent by researchers exploring the relation between the fracture toughness of soft polymer gels characterized with traditional testing methods and the inherent material properties related to crack growth [10,11,28–30], limited attention has been given to the critical force and energy involved in the initiation of a crack in soft solids. Understanding the mechanics behind crack initiation is not only vital to advance the current scientific knowledge on soft materials fracture but also relevant to numerous healthcare applications such as percutaneous needle insertion (biopsies, blood sampling, and anesthesia) [31]; robot-assisted surgeries [32,33], and design of advanced surgical instruments. Recently, Fakhouri et al. [34] studied the large strain deformation response and failure of chemically and physically cross-linked soft polymer gels with flat-end and spherically tipped indenters. Large strain response was characterized by a parameter $k'E$, where k' is an empirical constant found to correlate well with elastic moduli measurements from standard bulk mechanical testing techniques, such as shear rheology, for acrylic triblock copolymer gel materials [34], and resilin-based hydrogels [35]. The point of puncture is marked by a sharp drop in force, and is referred to as the critical force for puncture, P_c Fig. 2(b). A transition was reported for fracture initiation between stress-limited and energy-limited regime at a length scale governed by the indenter tip radius, R.

Due to similarities in fracture mechanics concepts between crack initiation associated with a puncture event and crack propagation, we hypothesized that critical puncture force is likely to be influenced by far-field compliance. Previous fundamental studies in adhesion and traditional fracture mechanics have revealed the influence of instrument stiffness on peak adherence force and stability of crack propagation [36,37]. In this paper, the effect of

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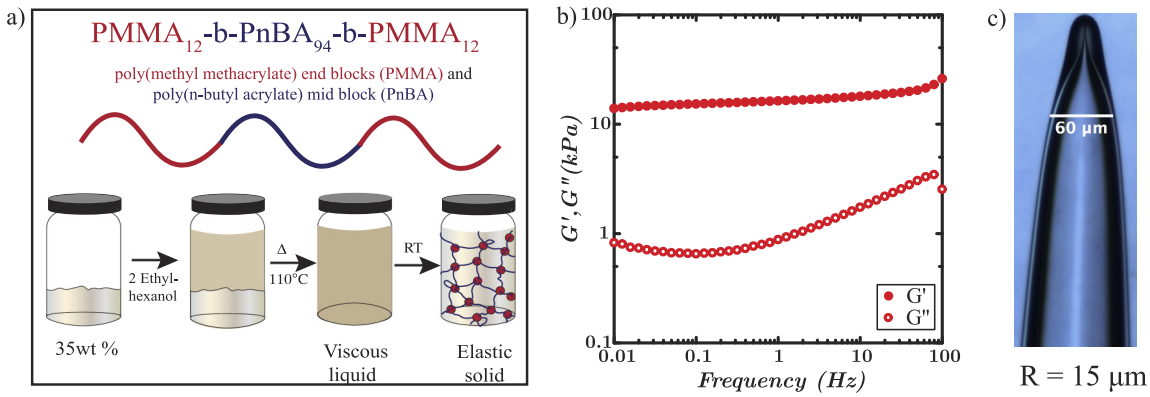

Fig. 1. (a) Formation of 35 wt% triblock copolymer gels by dissolving desired weight of copolymer pellets in 2 Ethyl-1-hexanol at 110 °C for 12 h to form a homogeneous melt. Homogeneous melt obtained is subsequently cooled gradually to form an elastic solid. Note that subscript numbers refer to each block's molecular weight in kg/mol. (b) Parallel-plate shear rheology of 35 wt% $PMMA_{12}$-$PnBA_{94}$-$PMMA_{12}$. The shear storage modulus, G', and the loss modulus, G'' are plotted versus frequency between 0.01 and 100 Hz. (c) Image of spherically-tipped indenter with radius R=15 μm. Indenters were formed by pulling glass capillaries with subsequent annealing in a microforge.

far-field compliance, C_s on P_c in highly elastic, acrylic triblock copolymer gels at a fixed polymer concentration and R is measured. We conducted a systematic set of experiments and varied both the ratio of C_s and effective gel compliance, C_g and global probe velocity, v_c over three orders of magnitude, in order to test our hypothesis. Our results demonstrate a significant impact of compliance on puncture mechanics, which can be understood by taking into account the change in local probe velocity, v_g, during puncture.

Puncture in soft materials is an important failure mechanism central to invasive medical procedures. In practicality, far-field compliance of a human or robotic arm can alter forces applied to the patient by cutting tools and surgical instruments [38].

2. Experimental

2.1. Materials

Acrylic triblock copolymer gels were chosen as model materials for this study due to their high elasticity upto large strains and thermoreversible gel transition [10,39,40]. ABA copolymers of poly(methyl methacrylate) end blocks (PMMA) and poly(n-butyl acrylate) mid block (PnBA) with molecular weights of 12 kg/mol and 94 kg/mol, respectively, were acquired from Kuraray Co. and used as received. The polymer gels are referred as $PMMA_{12}$-b-$PnBA_{94}$-b-$PMMA_{12}$, where subscript numbers refer to each block's molecular weight in kg/mol. Gels were formulated by dissolving copolymer pellets at 35 wt% in a mid-block selective solvent at low temperatures, 2-Ethyl-1-hexanol (purchased from Sigma-Aldrich) at 110 °C for 12 h in 20 mL glass vials. The homogeneous melt was subsequently cooled to room temperature, approximately 22 °C to form elastic gels as shown in Fig. 1(a). The mechanism for the elasticity of these gels is the aggregation of end-blocks into glassy domains (indicated by red circles in Fig. 1(a) on cooling below the critical micelle temperature and glass transition temperature of the endblocks [39,41]. The rubbery midblock segments form bridges between the endblock domains as indicated by blue strands in Fig. 1(a). Thus, these materials are physically associating ABA type triblock gels. The same thermal treatment was applied between puncture experiments to erase damage history.

Spherically tipped indenters were fabricated by pulling glass capillary tubes in a Narishige PC-10 and subsequently annealing the tips in a microforge. Spherical profiles resulted from surface tension and were confirmed by optical microscopy as shown in Fig. 1(c). In this study, we fabricated a spherically tipped indenter of radius, R=15 μm.

2.2. Methods

2.2.1. Design of an ultra-soft system to study effect of far-field compliance on puncture mechanics in gels.

A commercial experimental set-up (Stable Microsystems TA.XT plus Texture analyzer) was used to measure the force, P and displacement of indenter in the gel or gel displacement, δ_g during puncture experiments in soft gels. Thin, long rectangular plastic beams were incorporated in series with the spherically-tipped indenter and the load cell of the instrument schematically depicted as a spring in Fig. 2(a). A 3D printed adaptor was fabricated to provide an interface for the spring beam with the load cell. The spring beams were inserted in thin slots provided in the vertical arms of the adaptor (simply supported beam, large, 5 mm diameter fixed supports). The indenters were then rigidly attached to the Luer lock connection in the center of the spring beam. A detailed figure of the experimental set-up is shown in Appendix Figure A.7(a). Far-field compliance, C_s, is defined as the compliance of the spring beam, which was tuned by using beams of varying thickness at a constant length providing a compliance range of C_s∼200-1000 μm/mN. The force-spring beam deflection (P-δ_s) response of indentation on a rigid substrate for the experimental set-up incorporating spring beams and the 3D printed part is shown in Appendix Figure A.7(b). The compliance of the set-up without the beams was measured to be of the order of 0.1 μm/mN (Appendix Figure A.7(b)), which is negligible in comparison to the compliance of spring beams, and hence will be taken as ∼0 for the present study. The exact measurement of the compliance was limited by the smallest force and displacement resolution of the instrument.

P-δ_g response for a puncture experiment conducted in a 35 wt% triblock copolymer gel with a spherically tipped indenter of radius, R=15 μm, in an infinitely stiff experimental set-up (with no spring beam) corresponding to C_s∼0 is shown in Fig. 2(b). Upon indentation, resistive forces increase as the indenter deforms the gel until puncture occurs at P_c.

The effective compliance at the gel–indenter interface is defined as C_g which is a function of E and geometry of the indenter. Fakhouri et al. [34] described the large strain deformation behavior of a soft gel indented by flat and spherically tipped indenters with: $P=k''ER\delta_g+k'E\delta_g^2$, where R is the indenter tip radius, k' is an empirical constant, and k'' is a constant depending upon indenter tip geometry, obtained from Hertzian contact mechanics for flat and spherically tipped indenters [42]. This deep indentation equation was used to fit the loading curve in Fig. 2(b), and to approximate gel compliance, $C_g = \left.\frac{d\delta_g}{dP}\right|_{\delta_g=\delta_{g,c}}$, where $\delta_{g,c}$ is the critical depth for puncture, by plotting the inverse derivative of the fitted curve with