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Seamless modulus gradient structures for highly resilient, stretchable system integration

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ABSTRACT

Hybrid system integration of rigid components into stretchable systems is often necessary when targeting for valuable functions in various scenarios. Among them, (Young's) modulus gradient structures for system integration demonstrate excellent mechanical performance when stretched. However, the mechanical reliability is still limited under large deformation due to the inherent interface between materials of different modulus. Here, a seamless transition between heterogeneous moduli parts made with polydimethylsiloxane (PDMS)-based elastomers is presented for stretchable system integration by simply tuning their modulus via introducing a small amount of an additive into some parts of the substrate. These gradient structures not only provide a high stretchability (~250%) for the overall system, but also improve the resilience of the system (can be stretched up to 50,000 cycles from 0 to 150% global strain) at the same time. The seamless modulus gradient structures provide a simple and effective way of allowing highly resilient and stretchable system integration for various soft intelligent systems.

1. Introduction

Offering unsurpassed morphological dynamics and adaptability, soft intelligent systems with excellent mechanical compliance and stretchability of materials or structures, such as soft electronics and robotics, have shown great advantages when interacting with humans or the environment [1–3]. A few are listed here, for instance, a conformal sensor array for high resolution brain signal mapping [4], a new balloon catheter with feedback signals [5], a soft gripper that can manipulate fragile, random, and complex shaped objects [6,7], a self-driven hopping robot that can explore in complex surroundings [8], a tissue-engineered artificial ray guided by light [9], wearable silicon electronics based on system-level integration using serpentine structure [10] and an ultrathin stretchable system using polydimethylsiloxane (PDMS) suction-cup/substrate of gradual modulus [11]. To provide above mentioned vivid functions and exploit great morphological dynamics and adaptability at the same time, heterogeneous material-made rigid components including functional materials, rigid components and their assembled systems in elastomer packaging, are often necessary when implementing soft systems in the practical situations.

Several strategies enabling hybrid system integration in elastomers have been reported, for instance, thin film-based island-bridge structures [12–15], or a hybrid structure with a localized stiff cell, for which a rigid component was embedded in a thicker elastomeric package [16]. Nevertheless, heterogeneous integration systems where heterogeneous modulus occurs cause stress concentration around integrated parts, and lead to high mechanical stress at the interface between rigid parts and soft substrates when stretched [17,18]. For engineering properties in general, functional graded materials has been an active research field for decades [19], to improve the structural integrity of components. The concept of modulus gradient structure is widely found in nature to achieve

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mechanical resilience [20–23], such as in the gradient interface tissue of living bodies [24]. Several modulus gradient structures with PDMS have been reported, either by depositing a diamond-like carbon film onto a silicone substrate through pulsed laser ablation of an isolated stiff circuit island [25], or by local tuning of mechanical properties of PDMS-made structures with a photo-inhibitor via UV light exposure [26–28]. However, the global stretchability of such systems has not been significantly improved, and delamination is a potential risk under large deformation. In addition, an out-of-plane modulus gradient structure was recently demonstrated [29].

As demonstrated above, when applying an external tensile force to a modulus gradient structure, such as an island-bridge design, different effective strains are induced in each region due to the large difference of engineering modulus between a stiff material and a soft material. The soft material region exhibits larger strain than that of the stiff region. Consequently, the resulted high stress concentration often arises at the interface between soft and stiff materials. Frequently, the interface between two materials is weaker and more prone to cracking, than the two materials independently. The stress concentration at the interface is typically higher if there is a large elastic mismatch between the two constituents [30]. Therefore, a seamless modulus gradient structure is effective to lower stress concentration and to achieve high stretchability as well as high resilience under a large deformation. Additionally, originating from the same material with very similar chemical properties, the seamless transition can act as a connection between the soft and stiff regions, thereby improving the interfacial strength and the overall stretchability.

By simply tuning modulus with a small amount of an additive (polyethylenimine ethoxylated, PEIE) in PDMS [31] and using it with a semi-cured state of PDMS [32], we present a seamless modulus gradient structure with different moduli parts, targeting highly resilient and stretchable system integration. The experimental results as well as numerical analysis for various modulus gradient structures indicate that the seamless modulus gradient greatly extends the overall stretchability compared to previous approaches, such as those in Refs. [25–28]. Furthermore, the integration of various rigid components, such as light-emitting diodes (LEDs), chips and batteries are integrated and demonstrated. It shows a stretchability up to a 250% strain, which is significantly higher than that made with homogeneous modulus PDMS (up to 80%). More importantly, a rigid component-integrated stretchable system shows great mechanical resilience under large deformation, which can be stretched over 50,000 load cycles from 0 to 150% global strain without any electrical or mechanical failures.

2. Results and discussion

As shown in Fig. 1a, we employed native PDMS (10:1 wt ratio of silicone base to curing agent in this paper) and modulus-tuned PDMS (hereafter, S3PDMS) [31] to obtain modulus gradient structures with a simple curing process. Since the S3PDMS was made from the same base of PDMS, with more compatible polymers next to one another, the base polymer chains in both PDMS and S3PDMS could be crosslinked at the interfaces and resulted in strong connections that can sustain large strain without breaking. Thus, a seamless connection between the S3PDMS and native PDMS without an obvious boundary, which is a seamless modulus gradient structure, was formed (Fig. S1).

To investigate the effective modulus and the effective strain of various combinations of different moduli, we tested samples of PDMS, S3P20, S3P30, S3P40, H20, H30 and H40 (S3P20, S3P30 and S3P40 are named for S3PDMS mixed with 20, 30 and 40 μl of PEIE, respectively. H20, H30 and H40 are named for seamless modulus gradient structures with PDMS parts and S3P20, S3P30 and S3P40 parts, respectively. The configurations of the samples are shown in Fig. S2). The maximum elongation and effective modulus of various elastomers and seamless modulus gradient structures without rigid components integration are shown in Fig. 1b. We can see that the modulus of PDMS (~2.0 MPa) is two orders of magnitude higher than the modulus of S3P40, which is ~24 kPa. Stretchability (maximum elongation at breaking) of PDMS (~100%) is less than that of S3PDMS (~300%). S3PDMS reveals both excellent mechanical stretchability and modulus tunability. Because of the stiff region (normal PDMS), the seamless modulus gradient structures (H20, H30 and H40) show smaller elongation at breaking than that of the pure ones (S3P20, S3P30 and S3P40, respectively), whereas the effective moduli are quite similar.

Moreover, we presented hybrid integration systems either with an LED or four rectifier chips having dip-type sharp connect legs (Fig. 1c and d). Both of them were stretched up to 150% under unidirectional strain and the PDMS part exhibited negligible deformation (~8%), compared to that of S3PDMS parts (~200%). The rigid components in stiff regions can be effectively protected regardless of shapes, numbers and integrated positions of the components.

To understand the stress/strain distribution of seamless modulus gradient structures, tensile tests of PDMS, S3P20, S3P30, S3P40, H20, H30 and H40 without rigid component integration were conducted (Fig. 2a and b). Because seamless modulus gradient structures included a small volume of stiff portion of PDMS, the effective moduli of H20, H30 and H40 were slightly higher than that of S3P20, S3P30 and S3P40, respectively. However, the maximum elongation of the former is smaller than that of the latter. This indicates that a hard part does not have a significant influence on compliance and stretchability of the whole system when the stiff region is small enough, compared to soft regions. Compared to the case of H40, the PDMS area between the S3P20 parts in H20 showed larger strain because the modulus difference of H20 is smaller than that of H40 (Fig. 2b).

Since the volumes are connected in series, with each part carrying the same average axial stress, a simple rule of mixtures model can be applied to estimate the overall stiffness. Considering the different moduli and volumes of a soft and a stiff region when the volume ratio of S3PDMS and PDMS is 2:1, the effective modulus of the seamless modulus gradient structure when stress is assumed homogeneous throughout the system can be expressed as [33].

$$\frac{V}{E_e} = \frac{2V_1}{E_1} + \frac{V_2}{E_2} \tag{1}$$

where E_e is the effective modulus of a modulus gradient structure, E_1 and E_2 are the moduli of the soft part and the hard part, respectively, and V_1 and V_2 are the volumes of the soft and hard part, respectively. The effective modulus can be derived as

$$E_e = \frac{E_1 E_2}{\frac{2V_1}{V}E_2 + \frac{V_2}{V}E_1} \tag{2}$$

The applied global strain is

$$\lambda_g = \frac{2\lambda_1 L_1 + \lambda_2 L_2}{2L_1 + L_2} \tag{3}$$

where L is the undeformed length, and λ is the stretch ratio. Subscripts 1 and 2 represent the soft and the hard part, respectively. By defining $\eta = L_1/L_2$, equation (3) can be expressed as