

1 Methods

1.1 Experimental

The SEPS and the LLDPE films are the same as used in our previous research on stretching-induced wrinkling of rubber/plastic laminate composites[1]. Bilayer laminate samples were prepared by bonding LLDPE films (either 120 μm thick or 50 μm thick), to SEPS rubber films using compression molding as follows. SEPS resin (kindly provided by Kraton Corp.) was compression molded into sheets of desired levels of thickness at 190°C. The LLDPE films, kindly provided by Mitsui Chemical Co., were then placed on top of the rubber sheets, using a few drops of methanol as a lubricating layer that also helped avoid blisters. The samples were left overnight to let the methanol to evaporate. The bilayer was then heated to 140°C (which is well above the melting temperature of LLDPE) under a small compressive load for 5 min to ensure bonding of the rubber and plastic.

Dog-bone shaped samples (6 mm width and a nominal gauge length of 20 mm) were cut from the resulting bilayer composite sheet using a punch. A thin layer of silicone grease was smeared onto the gauge section of the dog-bone shaped samples to render them tacky. Small black particles were then stuck onto this surface to serve as markers for Digital Image Correlation (DIC) video analysis. Tensile testing was conducted at a crosshead speed of 120 mm/min using an Instron UTM model 5565. Applied load was measured using a 50lbf load cell (Honeywell, model:31) The clamp-to-clamp distance of the dog-bone shaped samples was 40 mm for all tests, which is twice the gauge length. Tensile tests were video-recorded so that they could be analyzed using digital correlation analysis (DIC) techniques to obtain the strain map over the sample. The two layers remained fully bonded to each other during tensile deformation (and indeed also after releasing, upon which the plastic layer showed intense wrinkling[1]).

We considered the possibility that during the bonding stage of the bilayer preparation, the thickness of the rubber and/or plastic layer might change since both layers are capable of flow at the bonding temperature. Accordingly, a region of the samples immediately adjacent to each dog-bone specimen was also cut, its thickness measured, and then immersed in toluene to fully dissolve the SEPS. Toluene does not affect the LLDPE, and hence the remaining LLDPE was removed from the solvent, dried, and its thickness measured; this corresponds to the actual thickness of the LLDPE in each dog-bone shaped bilayer specimen. Typically, the thickness of the individual layers deviated no more than 10% from the nominal thickness. In all the data analysis, the actual measured thickness, and not the nominal values were used.

Tensile stretching experiments were also conducted on the SEPS and the LLDPE individually. For the SEPS, dog-bone shaped specimens were directly cut out of the compression-molded sheets. For LLDPE, it is well-known that plastic films (due to their manufacturing process) have significant residual stress and orientation, and hence anisotropy in their mechanical properties. Obviously, the bilayer samples, being heated to above the melting temperature of LLDPE, are not expected to have residual orientation. To obtain LLDPE specimens that are more comparable to the LLDPE in the bilayers, the following procedure was adopted. Films of the LLDPE were placed on a smooth stainless-steel sheet covered with a layer of liquid polyethylene glycol. These sheets were then placed on a heated platen held at 150°C for 4 minutes (adequate to melt the LLDPE) which was then observed to shrink along one direction. The stainless-steel sheet was then removed from the platen, and the sample was cooled. During this entire process, the polyethylene glycol served as an inert lubricant, preventing the LLDPE from sticking to the steel sheet. Dog-bone shaped specimens were then cut from this LLDPE sheet.

1.2 Stretch mapping by DIC

Let \mathbf{X} be the position vector of the integration point (ξ) of any element in the initial configuration. Also, let \mathbf{x} be the position vector of the same point in any deformed configuration. Then, the position vector can be described as a function of nodal position vectors using isoperimetric interpolation functions $N_I(\xi)$ commonly called as shape functions as,

$$\mathbf{X} = \sum_{I=1}^3 N_I(\xi) \mathbf{X}_I \quad (1)$$

$$\mathbf{x} = \sum_{I=1}^3 N_I(\xi) \mathbf{x}_I \quad (2)$$

Further, the deformation gradient, \mathbf{F} of an element can be evaluated at the integration point (ξ) by,

$$\mathbf{F} = \mathbf{j} \mathbf{J}^{-1} \quad (3)$$

Where,

$$\mathbf{j} = \text{Grad}_\xi \mathbf{x} = \sum_{I=1}^3 \mathbf{x}_I \otimes \nabla_\xi N_I \quad (4)$$

$$\mathbf{J} = \text{Grad}_\xi \mathbf{X} = \sum_{I=1}^3 \mathbf{X}_I \otimes \nabla_\xi N_I \quad (5)$$

Then Right Cauchy – Green deformation tensor, \mathbf{C} is evaluated as

$$\mathbf{C} = \mathbf{F}^T \mathbf{F} \quad (6)$$

Now the stretch along any direction \mathbf{m} can be evaluated by,

$$\lambda_m = \sqrt{\mathbf{m} \cdot \mathbf{C} \mathbf{m}} \quad (7)$$

When $\mathbf{m} = \mathbf{e}_1$, the axial direction of the specimen, the axial stretch,

$$\lambda_{11} = \sqrt{C_{11}} \quad (8)$$

2 Results

2.1 Mechanical behavior of LLDPE/SEPS bilayer

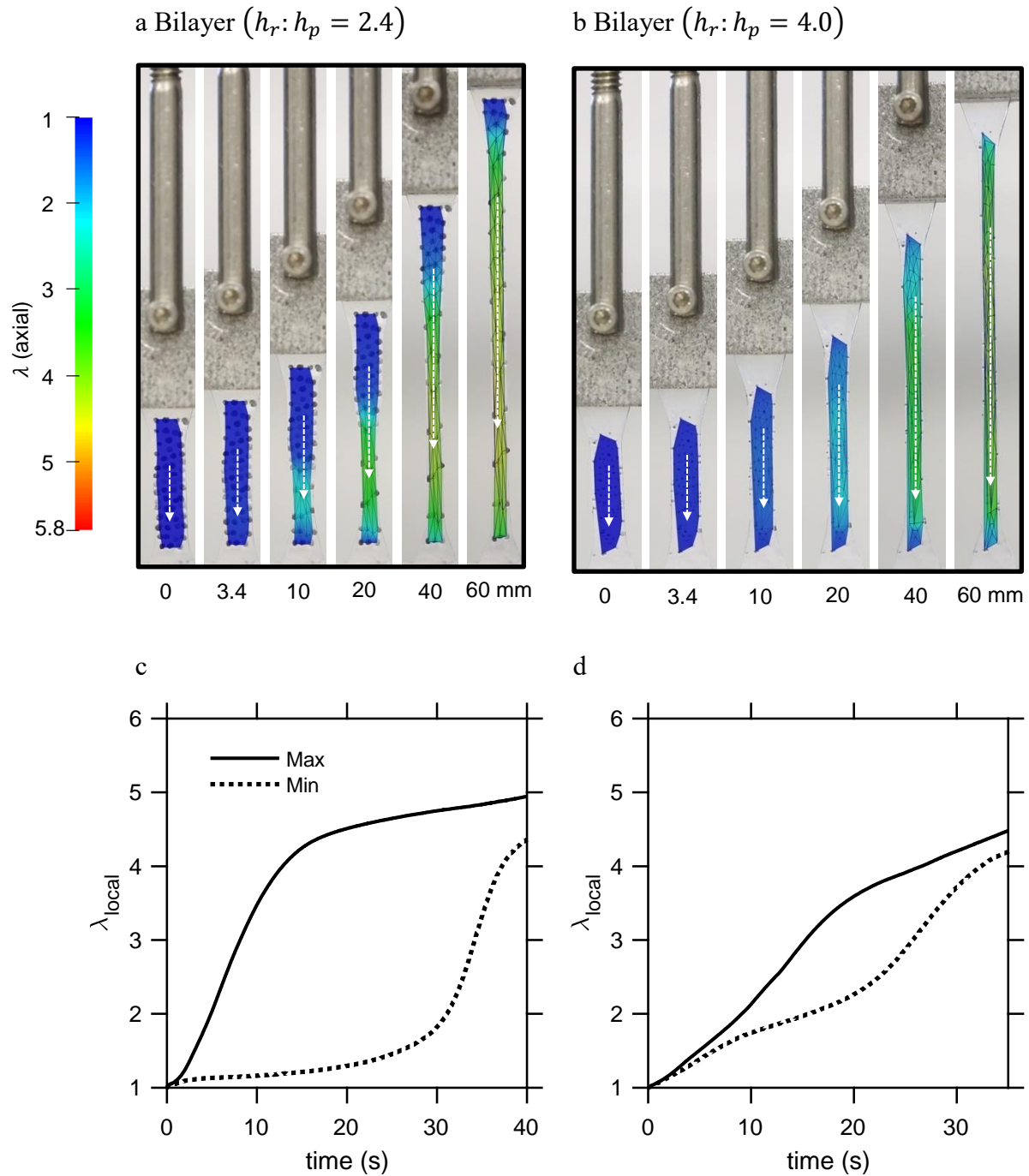


Figure S1: Stretch map for bilayers with rubber:plastic thickness ratio of 2.4 (a) and 4.0 (b). Stretch maps are superimposed on the corresponding configuration of deformation in each frame at crosshead

displacements noted below each frame. (c,d) Maximum stretch in the ROI (white dotted line in (a,b)) and the minimum stretch in the ROI is plotted against time of deformation for bilayers shown in (a,b) respectively.

3 Discussion

3.1 Inhomogeneity plot

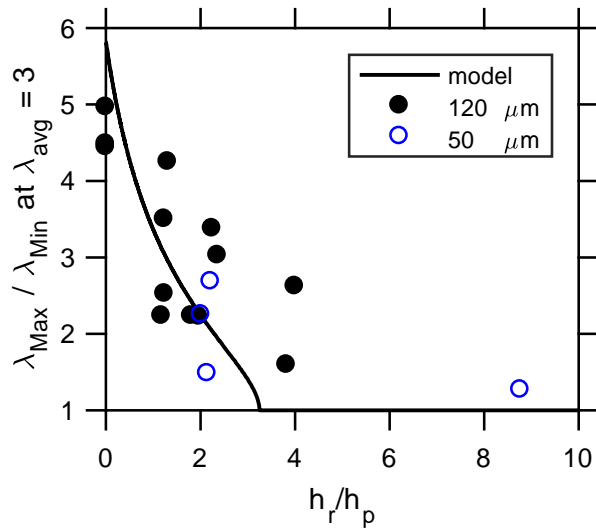


Figure S2: The ratio of maximum stretch in the ROI to the min stretch is plotted for freestanding LLDPE and for bilayers of rubber:plastic thickness ratio of 1-9, when the average stretch in the ROI is 3.

References

1. Yang, J., et al., *Stretching-induced wrinkling in plastic-rubber composites*. *Soft Matter*, 2017. **13**(4): p. 776-787.