Supporting Information

Stepped Moduli in Layered Composites

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• Mechanical properties of latex, PE, and Kevlar sheets

To investigate the contribution of each material when it is combined into the steppedmodulus composites, we first measured the Young's modulus of single sheets of latex, PE and Kevlar by performing a tensile test. We prepared strips (3 cm in width, $W \times 15$ cm in length, L) of latex, polyethylene (PE) (purchased from McMaster Carr) and Kevlar (purchased from Fibre Glast). We tested the mechanical properties of each strip using a tensile tester (Instron 5566) by measuring stress while applying constantly increasing strain with a rate of 60 mm/min. The tested area of each sample was 3 cm (W) \times 9 cm (L).



Figure S1. Force vs. extension and stress vs. strain curves of Kevlar, PE and latex strips. The size of the strips tested was 30 mm in width and 90 mm in length. The Young's modulus of the three materials differed by four orders of magnitude (2.7 MPa, 55 MPa and 24 GPa).

• Stepped modulus composites composed of thermoplastic polyurethane and a silicone based elastomer



Figure S2. Force vs. extension curves of a silicone-based elastomer (Ecoflex), thermoplastic polyurethane (TPU), and a TPU-Ecoflex stepped modulus composite. The length difference of the strips of TPU and Ecoflex of the composite was 25 mm. The measured Young's moduli of Ecoflex and TPU are 44.2 kPa and 17.7 MPa.

• Mechanical Properties of a layered composite composed of materials with low elongation-at-break.



Figure S3. (A) Layered composites of four sheets of aluminium and a layer of latex at rest (top) and in tensile strain (bottom). (B) The force-extension of the composite shows sharp peaks at four threshold extensions that are 10, 20, 30 and 40 mm as the Al layers break from the shortest to the longest. The latex layer helps the shortest Al layer maintain its wavy configuration at rest and prevents the composite from totally separated into two pieces even after the longest Al layer breaks.

 Mathematical analysis of the stepped-modulus composites with different amplitudes To prove that the amplitude of the PE layer of the composites in sinusoidal configuration decreases along with the wavelength, we compare the arc lengths of two sine curves, ¹/₂ sin 2x and sin x from 0 to 2π, and show their arc lengths are equal to each other.

First, we derive a general equation to calculate the arc length of an arbitrary function, y = f(x), from point A to B which we define as *L*. ΔL is the infinitesimal arc length when A and B are infinitely close to each other (Figure S3 (A)) and it is described in equation (S1) from the Pythagorean Theorem,.

$$(\Delta L)^2 = (\Delta x)^2 + (\Delta y)^2$$
(S1)

In a derivative form, the equation (S1) can be divided by dx, then *L* can be described as a function of integral shown in equation (S3).

$$\left(\frac{dL}{dx}\right)^2 = \left(\frac{dx}{dx}\right)^2 + \left(\frac{dy}{dx}\right)^2 = 1 + \left(\frac{df(x)}{dx}\right)^2$$
(S2)
$$\therefore L = \int \sqrt{1 + \left(\frac{df(x)}{dx}\right)^2} dx$$
(S3)

By using the equation (S3), we compare the arc lengths of two sine curves, $\frac{1}{2} \sin 2x$ and $\sin x$ from 0 to 2π . First, in the case of $\sin x$, the arc length from 0 to 2π is described in the equations below.

$$L = \int_{0}^{2\pi} \sqrt{1 + (\cos x)^2} \, dx \tag{S4}$$

$$=4\int_{0}^{\frac{\pi}{2}}\sqrt{1+(\cos x)^{2}}dx$$
 (S5)

$$=4\int_{0}^{\frac{\pi}{2}}\sqrt{2-(\sin x)^{2}}dx$$
 (S6)

$$=4\sqrt{2}\int_{0}^{\frac{\pi}{2}}\sqrt{1-\frac{1}{2}(\sin x)^{2}}dx$$
(S7)

Here, the integral term can be replaced by the complete elliptic integral of the second kind,

$$E(m^2) = \int_{0}^{\frac{\pi}{2}} \sqrt{1 - m(\sin u)^2} \, du$$
. Then the arc length is $4\sqrt{2}E(\sqrt{\frac{1}{2}}) \approx 7.64$.

Second, we calculate the arc length of $\frac{1}{2}\sin 2x$ from 0 to 2π to compare it with that of $\sin x$.

$$L = \int_{0}^{2\pi} \sqrt{1 + (\cos 2x)^2} \, dx \tag{S8}$$

We define θ as 2x to change the variable, then $dx = \frac{1}{2}d\theta$ and the equation (S8) can be replaced with (S9).

$$= \frac{1}{2} \int_{0}^{4\pi} \sqrt{1 + (\cos \theta)^2} d\theta$$
(S9)
$$= \int_{0}^{2\pi} \sqrt{1 + (\cos \theta)^2} d\theta$$
(S10)

The equation (S10) is equal to the equation (S4), thus the arc length of $\frac{1}{2}\sin 2x$ from 0 to 2π is

also $4\sqrt{2}E(\sqrt{\frac{1}{2}}) \approx 7.64$.



Figure S4. (A) The arc length of an arbitrary function, f(x) from A to B can be described as an equation using the Pythagorean Theorem. (B) The amplitude of a longer sheet (red) decreases proportionally to its wavelength (one third in this exemplary diagram) when the total arc length is constant.

• Stepped modulus composite with non-uniform amplitude of the stiff layer



Figure S5. (A) Four states of a stepped modulus composite with three sections of different arc length when the applied strain increases. Until the state D is reached, only the latex sheet stretches. (B) Force vs. extension curve of a latex-PE stepped modulus composite described in (A). The threshold extension is 36 mm which is equal to the sum of the length difference of three sections between the two sheets (10 mm + 12 mm + 14 mm).

• Stepped modulus composite with non-uniform wavelength of the stiff layer



Figure S6. Photographs of a PE-latex composite with non-uniform wavelength of the PE layer at top ((A) and (C)) and perspective views ((B) and (D)) on a grid. The composite is at rest in (A) and (B) and in tensile strain in (C) and (D). The red lines represent the attachment of the PE and latex layers, and the blue dotted lines in (B) and (D) represent the shape of the PE layer. The gradations on the grid are 1 mm and the black line shows the half of the length of the composite. When the composite is stretched from L_{Latex} (60 mm) to $L_{Latex} + \Delta L$ (90 mm), the composite distorts in the direction of its width.

• Composites with rod-coil configuration



Figure S7. (A) Photograph of a Kevlar-TPU-Ecoflex composite with rod-coil configuration. The TPU strip was wrapped with Kevlar fibers in a helical fashion; this TPU-Kevlar composite was wrapped around a rod of Ecoflex. (B) Force-extension profiles of a Kevlar-TPU-Ecoflex (\Box) and a TPU-Ecoflex (Δ) composites, and a single rod of Ecoflex (\circ).

• Composite with embedded configuration



Figure S8. (A) Photograph of a Kevlar-TPU-Ecoflex composite with embedded configuration. The Kevlar-TPU composite was embedded in Ecoflex in a sinusoidal configuration. (B) Forceextension profiles of the composite. The composite delaminated after ~ 60 mm of extension. (C, D) Simulated stress distribution in model geometries of Kevlar-Ecoflex composites in embedded (C) and layered (D) configurations in tension. The length and thickness of the Ecoflex in the model geometries are 75.4 mm and 7 mm. The amplitude and period of Kevlar is 5 mm and 25 mm. For simplification, the tensile load (10 N) is assumed to be applied only on the right side of the composite.