Fabrication, characterisation and modelling of uniform and gradient auxetic foam sheets

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Supplementary Material

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Analytical expressions for the on-axis and off-axis properties of a 2D hexagonal honeycomb.

Consider the honeycomb cell geometry shown in Fig. S1 (and also shown in the schematic inserts in Figure 7a).

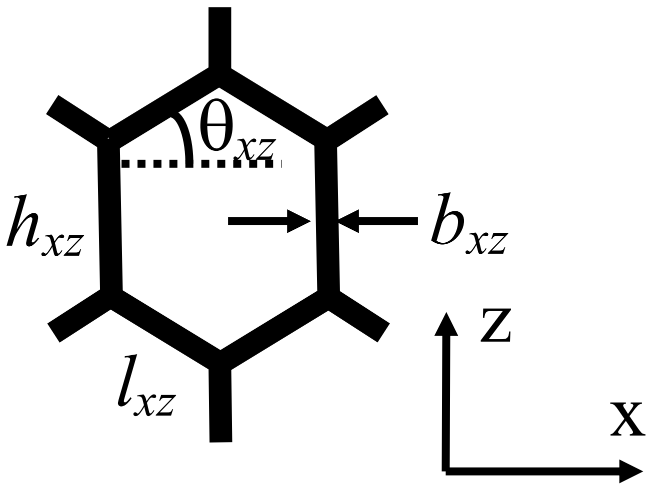

Figure S1. Hexagonal honeycomb cell geometrical parameters (x-z plane)

The Poisson's ratio and Young's modulus expressions for the honeycomb based on the cell geometry shown in Fig. S1 are adapted from Masters and Evans [35] here:

$$\nu_{xz} = \frac{\sin\theta_{xz}\cos^2\theta_{xz}\left(\frac{1}{K_{hf}} - \frac{1}{K_s}\right)}{\left(\frac{h_{xz}}{l_{xz}} + \sin\theta_{xz}\right)\left[\frac{\sin^2\theta_{xz}}{K_{hf}} + \frac{\cos^2\theta_{xz}}{K_s}\right]} \tag{S1}$$

$$\nu_{zx} = \frac{\sin\theta_{xz}\left(\frac{h_{xz}}{l_{xz}} + \sin\theta_{xz}\right)\left(\frac{1}{K_{hf}} - \frac{1}{K_s}\right)}{\left[\frac{\cos^2\theta_{xz}}{K_{hf}} + \frac{2\frac{h_{xz}}{l_{xz}} + \sin^2\theta_{xz}}{K_s}\right]} \tag{S2}$$

$$E_x = \frac{\cos\theta_{xz}}{b_{xz}\left(\frac{h_{xz}}{l_{xz}} + \sin\theta_{xz}\right)\left[\frac{\sin^2\theta_{xz}}{K_{hf}} + \frac{\cos^2\theta_{xz}}{K_s}\right]} \tag{S3}$$

$$E_z = \frac{\left(\frac{h_{xz}}{l_{xz}} + \sin\theta_{xz}\right)}{b_{xz}\cos\theta_{xz}\left[\frac{\cos^2\theta_{xz}}{K_{hf}} + \frac{2\frac{h_{xz}}{l_{xz}} + \sin^2\theta_{xz}}{K_s}\right]} \tag{S4}$$

where h and l are ‘vertical’ (aligned along z) and ‘oblique’ honeycomb rib lengths, respectively, θ is the angle of the oblique ribs with the horizontal (x) axis, b is the rib thickness, and the subscript ‘xz’ applied to the geometrical parameters denotes they are in the x-z plane. K_s is a force constant governing the rib stretching mode of deformation, and K_{hf} is a combination of the hinging (K_h) and flexure (K_f) force constants:

$$K_{hf} = \frac{K_h K_f}{K_h + K_f} \tag{S5}$$

The shear modulus G_{xz} expression is derived from the shear moduli expressions for each of the individual modes of deformation as follows:

$$\frac{1}{G_{xz}} = \frac{1}{G_f} + \frac{1}{G_h} + \frac{1}{G_s} \tag{S6}$$

where G_f, G_h and G_s are the rib flexure, hinging and stretching shear moduli, giving

$$G_{xz} = \left[\frac{b_{xz}\left(\frac{h_{xz}}{l_{xz}}\right)^2\left(1+\frac{2h_{xz}}{l_{xz}}\right)\cos\theta_{xz}}{K_{hf}\left(\frac{h_{xz}}{l_{xz}}+\sin\theta_{xz}\right)} + \frac{b_{xz}\left(1+\frac{h_{xz}}{l_{xz}}\sin\theta_{xz}\right)^2}{K_s\cos\theta_{xz}\left(\frac{h_{xz}}{l_{xz}}+\sin\theta_{xz}\right)} \right]^{-1} \tag{S7}$$

In (S7), the first term on the right hand side employs the expression for shear modulus due to rib flexure from Gibson and Ashby [19], extended to include rib hinging by acknowledging that the components of applied force causing flexure and hinging, and rib end-to-end displacement directions, are the same for both mechanisms (hence the same individual Poisson's ratio expressions, and Young's moduli expressions which differ only by the respective force constants). The second term on the right hand side of (S7) is the expression for shear modulus due to rib stretching and is taken from Masters and Evans [35].

The expression for off-axis Poisson's ratio response is again adapted from [35]:

$$\nu_{xz}(\phi) = E_x(\phi)\left[\frac{(\cos^4\phi+\sin^4\phi)\nu_{xz}}{E_x} - \cos^2\phi\sin^2\phi\left(\frac{1}{E_x}+\frac{1}{E_z}-\frac{1}{G_{xz}}\right)\right] \tag{S8}$$

where

$$E_x(\phi) = \left[\frac{\cos^4\phi}{E_x} + \cos^2\phi\sin^2\phi\left(\frac{1}{G_{xz}}-\frac{2\nu_{xz}}{E_x}\right)+\frac{\sin^4\phi}{E_z}\right]^{-1} \tag{S9}$$

Analytical expressions for strain in a 2D hexagonal honeycomb deforming by combined flexing, hinging and stretching of cell ribs.

To predict variation of the mechanical properties with loading strain, we first determine the generic off-axis strain from the two on-axis components:

$$\varepsilon_x(\phi) = \varepsilon_x\cos^2\phi + \varepsilon_z\sin^2\phi \tag{S10}$$

where

$$\varepsilon_x = \ln\left(\frac{X}{X_0}\right) = \ln\left(\frac{l_{xz}\cos\theta_{xz}-\delta_{xz}\sin\theta_{xz}}{l_{xz(0)}\cos\theta_{xz(0)}-\delta_{xz(0)}\sin\theta_{xz(0)}}\right) \tag{S11}$$

$$\varepsilon_z = \ln\left(\frac{Z}{Z_0}\right) = \ln\left(\frac{h_{xz}+l_{xz}\sin\theta_{xz} + \delta_{xz}\cos\theta_{xz}}{h_{xz(0)}+l_{xz(0)}\sin\theta_{xz(0)} + \delta_{xz(0)}\cos\theta_{xz(0)}}\right) \quad \text{(S12)}$$

and X and Z are the unit-cell lengths along the x and z directions, respectively, δ is the deflection of the beam due to flexing [19], and the subscript '0' denotes the initial (undeformed) value of the associated geometrical parameter. Substituting (S11) and (S12) into (S10), and assuming $\delta_{xz(0)}$ = 0, yields (1) in the manuscript.

Consider, firstly, a load applied parallel to the x direction. In this case, there is no component of the applied load to cause axial extension of the ribs of length h_{xz} and so $h_{xz} = h_{xz(0)}$. The variation of oblique rib length l_{xz} with oblique rib angle θ_{xz} under an x-directed load was derived using a similar approach to that used in the Nodule-Fibril model for microporous polymers [37]. Consider the forces acting on a rib of length l_{xz} subject to loading along the x direction for unit thickness in the y direction normal to the x-z plane – Figure S2. The change in the moment applied to each rib of length l_{xz} in this case is given by

$$\Delta M = -\frac{\Delta\sigma_x Z l_{xz} sin\theta_{xz}}{2} \quad \text{(S13)}$$

where $\Delta\sigma_x$ is the change in stress applied in the x direction.

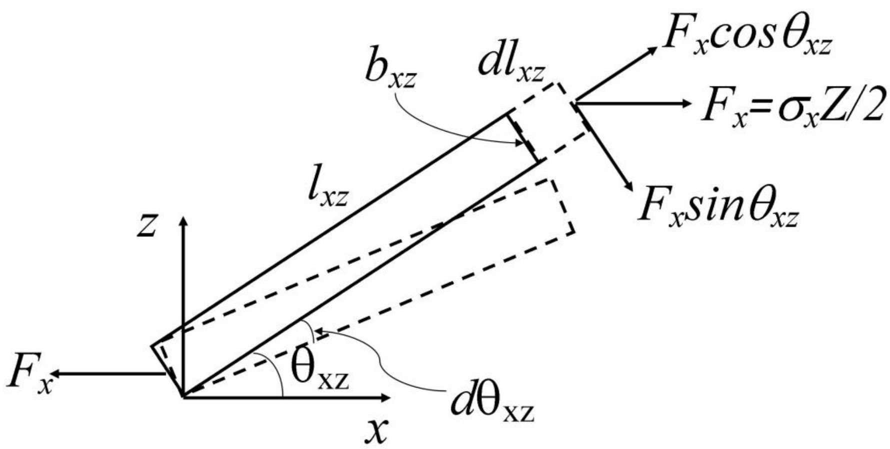

Figure S2 – Forces acting on rib of length l_{xz} subject to a load applied along the x direction.

The hinging force constant is defined by

$$K_h = \Delta M / \Delta\theta_{xz} \qquad \text{(S14)}$$

where $\Delta\theta_{xz}$ is the change in rib angle due to the change in moment. Substituting (S13) into (S14) allows the change in angle (amount of rib hinging) to be related to the change in applied stress:

$$\Delta\theta_{xz} = -\frac{\Delta\sigma_x Z l_{xz} sin\theta_{xz}}{2K_h} \qquad \text{(S15)}$$

To determine the amount of rib stretching due to the change in applied stress, first consider the change in force applied along the length of the rib:

$$\Delta F = \frac{\Delta\sigma_x Z cos\theta_{xz}}{2} \qquad \text{(S16)}$$

The stretching force constant relates the change in rib length, Δl_{xz}, to the change in force ΔF along the length of the rib:

$$K_s = \Delta F / \Delta l_{xz} \qquad \text{(S17)}$$

Substituting (S16) into (S17) allows the change in rib length to be related to the change in applied stress:

$$\Delta l_{xz} = \frac{\Delta\sigma_x Z cos\theta_{xz}}{2K_s} \qquad \text{(S18)}$$

In the limit of infinitesimal changes in rib angle and length, (S15) and (S18) yield

$$\frac{dl_{xz}}{d\theta_{xz}} = -\frac{K_h cot\theta_{xz}}{l_{xz} K_s} \qquad \text{(S19)}$$

Rearranging (S19) and integrating with the assumption that K_h/K_s remains constant:

$$\int_{l_{xz(0)}}^{l_{xz}} l_{xz} dl_{xz} = -\frac{K_h}{K_s} \int_{\theta_{xz(0)}}^{\theta_{xz}} cot\theta_{xz} d\theta_{xz} \qquad \text{(S20)}$$

giving

$$l_{xz}^2 = -\frac{2K_h}{K_s} \ln\left(\frac{sin\theta_{xz}}{sin\theta_{xz(0)}}\right) + l_{xz(0)}^2 \qquad \text{(2)}$$

To determine the amount of rib flexing due to the change in applied stress, first consider the flexure force constant defined by

$$K_f = \Delta F/\Delta\delta_{xz} \tag{S21}$$

where $\Delta\delta_{xz}$ is the change in rib deflection due to the change in moment. The change in the force applied perpendicular to the end of each rib of length l_{xz}, causing flexure, in this case is given by

$$\Delta F = -\frac{\Delta\sigma_x Z sin\theta_{xz}}{2} \tag{S22}$$

Substituting (S22) into (S21) allows the change in deflection (amount of rib flexing) to be related to the change in applied stress:

$$\Delta\delta_{xz} = -\frac{\Delta\sigma_x Z sin\theta_{xz}}{2K_f} \tag{S23}$$

In the limit of infinitesimal changes in rib angle and length, (S15) and (S23) yield

$$\frac{d\delta_{xz}}{d\theta_{xz}} = \frac{K_h}{l_{xz}K_f} \tag{S24}$$

Rearranging (S24) and integrating with the assumption that K_h/K_f remains constant:

$$\int_{\delta_{xz(0)}}^{\delta_{xz}} d\delta_{xz} = \frac{K_h}{l_{xz}K_f}\int_{\theta_{xz(0)}}^{\theta_{xz}} d\theta_{xz} \tag{S25}$$

giving

$$\delta_{xz} = \frac{K_h}{K_f}\frac{(\theta_{xz}-\theta_{xz(0)})}{l_{xz}} + \delta_{xz(0)} \tag{3}$$

Consider now loading along the z direction. The change in the moment applied to each rib of length l_{xz} in this case is given by

$$\Delta M = \frac{\Delta\sigma_z X l_{xz} cos\theta_{xz}}{2} \tag{S26}$$

where $\Delta\sigma_z$ is the change in stress applied in the z direction.

Substituting (S26) into (S14) allows the change in angle (amount of rib hinging) to be related to the change in applied stress:

$$\Delta\theta_{xz} = \frac{\Delta\sigma_z X l_{xz} \cos\theta_{xz}}{2K_h} \tag{S27}$$

The change in force applied along the length of the rib of length l_{xz}:

$$\Delta F = \frac{\Delta\sigma_z X \sin\theta_{xz}}{2} \tag{S28}$$

Substituting (S28) into (S17) allows the change in rib length to be related to the change in applied stress:

$$\Delta l_{xz} = \frac{\Delta\sigma_z X \sin\theta_{xz}}{2K_s} \tag{S29}$$

In the limit of infinitesimal changes in rib angle and length, (S27) and (S29) yield

$$\frac{dl_{xz}}{d\theta_{xz}} = \frac{K_h \tan\theta_{xz}}{l_{xz} K_s} \tag{S30}$$

Rearranging (S30) and integrating with the assumption that K_h/K_s remains constant:

$$\int_{l_{xz(0)}}^{l_{xz}} l_{xz} dl_{xz} = \frac{K_h}{K_s} \int_{\theta_{xz(0)}}^{\theta_{xz}} \tan\theta_{xz}\, d\theta_{xz} \tag{S31}$$

giving

$$l_{xz}^2 = \frac{2K_h}{K_s} \ln\left(\frac{\cos\theta_{xz(0)}}{\cos\theta_{xz}}\right) + l_{xz(0)}^2 \tag{S32}$$

For loading along z it is also necessary to consider stretching of ribs of length h_{xz}. The change in force applied along the length of the rib of length h_{xz}:

$$\Delta F = \Delta\sigma_z X \tag{S33}$$

The stretching force constant relating the change in rib length, Δh_{xz}, to the change in force ΔF along the length of the rib:

$$K_s^{h_{xz}} = \frac{\Delta F}{\Delta h_{xz}} \tag{S34}$$

Substituting (S33) into (S34) allows the change in rib length to be related to the change in applied stress:

$$\Delta h_{xz} = \frac{\Delta\sigma_z X}{K_s^{h_{xz}}} = \frac{\Delta\sigma_z X}{K_s}\frac{h_{xz}}{l_{xz}} \tag{S35}$$

where we have used the relationship between the stretching force constants for both rib types assuming they are both made of the same elastic material (see (S41) and (S41a) below). In the limit of infinitesimal changes in rib angle and length, (S27) and (S35) yield

$$\frac{dh_{xz}}{d\theta_{xz}} = \frac{2h_{xz}K_h}{l_{xz}^2 \cos\theta_{xz} K_s} \tag{S36}$$

Rearranging (S36) and integrating with the assumption that K_h/K_s remains constant:

$$\int_{h_{xz(0)}}^{h_{xz}} \frac{dh_{xz}}{h_{xz}} = \frac{2K_h}{l_{xz}^2 K_s}\int_{\theta_{xz(0)}}^{\theta_{xz}} \sec\theta_{xz}\, d\theta_{xz} \tag{S37}$$

giving

$$h_{xz} = h_{xz(0)} exp\left[\frac{2K_h}{l_{xz}^2 K_s}\ln\left(\frac{\sec\theta_{xz} + \tan\theta_{xz}}{\sec\theta_{xz(0)} + \tan\theta_{xz(0)}}\right)\right] \tag{S38}$$

The change in the force applied perpendicular to the end of each rib of length l_{xz}, causing flexure, for z-directed loading is given by

$$\Delta F = \frac{\Delta\sigma_z X \cos\theta_{xz}}{2} \tag{S39}$$

Substituting (S39) into (S21) allows the change in deflection (amount of rib flexing) to be related to the change in applied stress:

$$\Delta\delta_{xz} = \frac{\Delta\sigma_z X \cos\theta_{xz}}{2K_f} \tag{S40}$$

In the limit of infinitesimal changes in rib angle and length, (S27) and (S40) yield (S24) and so the deflection due to flexing as a function of rib angle change (due to hinging) under z-directed loading is the same as for x-directed loading and is given by (3).

Force constants assuming elastic cell rib material.

Expressions relating the force constants defined in (S14), (S17), (S21) and (S34) to material properties and rib geometry have been developed previously [35]:

$$K_s = \frac{E_s wt}{l} \tag{S41}$$

$$K_s^{h_{xz}} = \frac{E_s wt}{h} = \frac{E_s wt}{l}\frac{l}{h} = K_s \frac{l}{h} \tag{S41a}$$

$$K_f = \frac{E_s wt^3}{l^3} \tag{S42}$$

$$K_h = \frac{G_s wt}{l} \qquad \text{(hinging via shearing of junction)} \tag{S43a}$$

$$K_h = \frac{E_s wt^3}{6l^2 q} \qquad \text{(hinging via local bending at junction)} \tag{S43b}$$

where E_s and G_s are the Young's and shear moduli of the rib material, w is the rib depth, and q is the axial length of the curved hinge.

Dividing (S5) by (S41) and substituting (S42) and (S43a), the expression for K_{hf}/K_s when hinging occurs via junction shearing is:

$$\frac{K_{hf}}{K_s} = \frac{1}{\left(\frac{l}{t}\right)^2 + \frac{E_s}{G_s}} \tag{S44a}$$

Similarly, the expression for K_{hf}/K_s when hinging occurs via junction bending is:

$$\frac{K_{hf}}{K_s} = \frac{t^2}{l^2 + 6lq} \tag{S44b}$$

The minimum value of (S44b) occurs when $q \rightarrow l$:

$$\frac{K_{hf}}{K_s} = \frac{t^2}{7l^2} \tag{S44c}$$

Rationale for choice of force constant values used in model predictions.

We arbitrarily use $t/l = 0.2$, which seems reasonable from the foam micrographs, and assume isotropic rib material having Poisson's ratio $\nu_s = 0.25$, such that $E_s/G_s = 2(1 + \nu_s) = 2.5$. Substituting these values into (S44a) and (S44c), we find the ballpark range for K_{hf}/K_s from 0.006 to 0.036 – i.e. order of magnitude difference between the upper and lower limits.

Given the approximations inherent in employing an idealised 2D model of regular cells to real 3D cellular system displaying a distribution of cell sizes and shapes then, for the purposes of the calculations in the manuscript, we employ $0.004 < K_{hf}/K_s < 0.04$ for the predictions of Poisson's ratio versus cell geometry (Fig. 7). For the prediction of the mechanical properties as a function of strain in the *x-z* plane, we consider the case of $K_{hf}/K_s = 0.004$ and from (S41) and (S42) use $K_f/K_s = (t/l)^2 = (0.2)^2 = 0.04$. Hence from (S5) we find $K_f/K_h = 9$ and $K_s/K_h = 225$.

For the *x-y* plane predictions as a function of strain, as discussed in the manuscript the 'stretching' mode is enhanced over that expected for an elastic 2D material due to the 3D nature and orientation of the ribs in/out of the *x-y* plane. Accordingly, K_s is reduced in the *x-y* plane predictions and a value of $K_s/K_h = 3$ was employed. Retaining $K_f/K_h = 9$ produces a value of $K_{hf}/K_s = 0.3$ in this case.