Supporting Information for "Structural hierarchy confers error tolerance in biological materials"

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Supporting Information (SI)

Finding the critical bond portions. Critical bond portions for a network with N levels are computed from simulation data for each level by choosing all points in bond portion space for which the network is fully connected on the other $N-1$ levels, and the stiffness is non-zero. For one and two levels, we then fit a plot of stiffness vs. the bond portion of interest to an equation of the form of Eq. 1 from the main text. For one-level networks, we sampled bond portions from 0.6 to 1.0 in increments of 0.2. For two level networks, we sampled bond portion space from 0.55 to 1.0 in increments of 0.05 along both the large-scale and small-scale directions. Owing to the computational expense of simulated three-level networks, we sampled bond portion space from 0.8 to 1.0 in increments of 0.1 for all levels. As 0.8 appeared to be below the critical bond portion for the small scale, we also simulated a network with bond portions of 1.0, 1.0 and 0.85 on the large, medium, and small scales, respectively, so as to have at least three points above the threshold of vanishing stiffness. From here, data for stiffness vs. bond portion for three level networks were fit to a line of the form

$$K(p) = a \cdot p + k_0 \qquad [S13]$$

and the x-intercept of this line was taken to be the critical bond portion. Our analysis is shown schematically in Fig. S1.

Analytical prediction of hierarchical robustness. First, we consider a nominal point far from any boundary in bond portion space. For N levels, we define an excess bond portion $p_{e,i}$ for each level, with $1 \leq i \leq N$:

$$p_{e,i} = p_i - p_{c,i} \qquad [S14]$$

Now let the deviation from the nominal excess bond on the ith level be δ_i, with each δ_i identically and independently distributed according to a distribution $\mathcal{P}_i$ with zero mean and standard deviation σ_i, such that the total probability distribution function for a set of displacements $\{\delta_1, \ldots, \delta_N\}$ is

$$\mathcal{P} = \prod_{i=1}^{N} \mathcal{P}_i(\delta_i) \qquad [S15]$$

Referring to Eq. 4 in the main text, we define the reduced stiffness

$$\bar{K} = \frac{K}{k \prod_{i=1}^{N} (1 - p_{c,i})} \qquad [S16]$$

With this definition we now find the expected deviation in the stiffness of a network with N hierarchical levels. The mean of $\bar{K}$ is

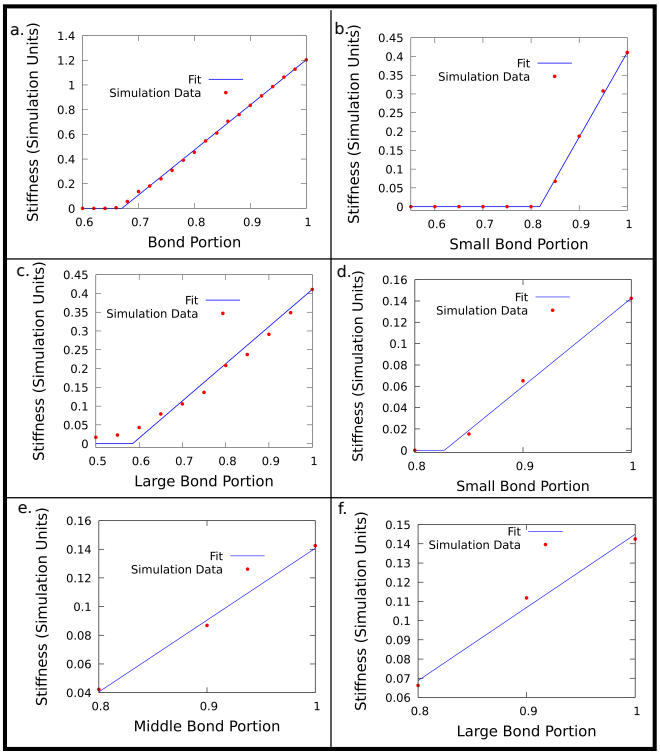

Fig. S1. Panel **a** illustrates the determination of the critical bond portion for one level. Panels **b** and **c** demonstrate the determination of the critical bond portion for the large and small-scale critical connectivities for the two-level case, and panels **d**-**e** illustrate determination of critical connectivity for the large, medium and small scales for the three-level case.

$$\left\langle \bar{K} \right\rangle = \int \cdots \int \left(\prod_{i=1}^{N} \left(p_{e,i} + \delta_i \right) \right) \mathcal{P}\left(\delta_1, \ldots, \delta_N \right) d\delta_1 \cdots d\delta_N \quad [S17]$$

$$= \prod_{i=1}^{N} p_{e,i} \quad [S18]$$

while the mean square stiffness is

$$\left\langle \bar{K}^2 \right\rangle = \int \cdots \int \left(\prod_{i=1}^{N} \left(\delta_i + p_{e,i} \right)^2 \right) \mathcal{P}\left(\delta_1, \cdots, \delta_N \right) d\delta_i \cdots d\delta_N \quad [S19]$$

$$= \prod_{i=1}^{N} \sigma_i^2 + p_{e,i}^2 \quad [S20]$$

The standard deviation in stiffness is then

$$\Delta \bar{K} = \sqrt{\prod_{i=1}^{N} \left(\sigma_i^2 + p_{e,i}^2 \right) - \prod_{i=1}^{N} p_{e,i}^2} \quad [S21]$$

Here it has been assumed that $p_{e,i} \gg \sigma_i$ for all i, so integration can be carried out with the assumption that Eq. 4 in the main text holds for all values δ_i that contribute appreciably to the integral. In the special case in which the excess bond portion is the same on each length scale, and each bond portion has the same standard deviation, Eq. (S21) reduces to

$$\sqrt{\left(\sigma^2 + p_e^2 \right)^N - p_e^{2N}}$$

, with $p_e = \bar{K}^{1/N}$. The relative error in stiffness then scales with N as

$$\frac{\Delta \bar{K}}{\bar{K}} = \frac{\sqrt{\left(\sigma^2 + p_e^2 \right)^N - p_e^{2N}}}{p_e^N} \quad [S22]$$

$$= \sqrt{\left(1 + \frac{\sigma^2}{p_e^2} \right)^N - 1} \quad [S23]$$

$$\approx \frac{\sqrt{N}\sigma}{p_e} \quad [S24]$$

or

$$\frac{\Delta \bar{K}}{\bar{K}} \approx \frac{\sqrt{N}\sigma}{\bar{K}^{1/N}} \quad [S25]$$

where the last approximation holds when $\sigma \ll p_e$. For a target $\bar{K}$, this functional form predicts the optimal number of levels to be

$$N^* = \lfloor -2 \ln\left(\bar{K} \right) \rfloor \quad [S26]$$

Accounting for Other Types of Error Distributions. As mentioned in the main text, we also accounted for two additional classes of distributions of random errors in assembly. In the first case, we still presume the errors to be independent on each scale and normally distributed, but we allow the standard deviation of error in bond portion at the smallest length scale to be different from the standard deviation for all other scales. For an N-level network, suppose $N-1$ levels exhibit random errors with standard deviation σ_a, while the remaining level exhibits random errors in bond portion with standard deviation σ_b. In this case, the probability distribution function for the N-dimensional vector of errors, $\vec{\delta}$, should take the form

$$\mathcal{P}\left(\vec{\delta} \right) = \frac{1}{\left(2\pi \right)^{2/N} \sigma_a^{N-1} \sigma_b} \exp\left\{ -\frac{1}{2\sigma_a^2} \prod_{i \neq 2} \delta_i^2 - \frac{\delta_2^2}{2\sigma_b^2} \right\} \quad [S27]$$

We consider once more a point in bond portion space with the same excess bond portion on each level, and that this excess bond portion is much greater than either σ_a or σ_b. A straightforward modification to the above derivation for a constant standard deviation yields

$$\frac{\bar{K}}{\Delta \bar{K}} = \sqrt{\frac{\sigma_b^2}{\bar{K}^{2/N}} + (N-1)\frac{\sigma_a^2}{\bar{K}^{2/N}} + (N-1)\frac{\sigma_a^2 \sigma_b^2}{\bar{K}^{4/N}}} \quad [S28]$$

We consider the greatest value for σ_b for an N-level network such that the relative variation in stiffness is no greater than the relative deviation in stiffness for a single-level network with a bond portion distribution of width σ_a. Equating the right-hand sides of Eq. (S25) and Eq. (S28) yields

$$\frac{\sigma_b}{\sigma_a} = \bar{K}^{2/N} \sqrt{\frac{1}{\bar{K}^2} - \frac{N-1}{\bar{K}^{2/n}}} \sqrt{\frac{1}{\bar{K}^{2/N} + (N-1)\sigma_a^2}} \quad [S29]$$

We show this behavior for the case in which the product of excess bond portions is fixed at 0.1, and at all levels of structure but the second, the standard deviation of the error in bond portion is 0.001 (Fig. S2a). The ratio of the maximum standard deviation on the second level such that the overall relative variation in stiffness remains less than or equal to that for a one-level network is plotted vs. the number of levels of hierarchy.

We can use the foregoing discussion to explore the marginal benefit of adding another level of hierarchy, by first considering a lattice with $N-1$ levels of structure, each of which has an identical distribution of bond portions with standard deviation σ_a. If this lattice, in turn, is used to construct each large-scale bond in an N-level lattice, we may identify the largest standard deviation of bond portion, σ_N, on the large scale such that the relative fluctuation in stiffness of the overall structure is not increased by the addition of another level. Note first that Eq. (S28) gives the relative fluctuation for the N-level lattice when σ_b is replaced by σ_N. Equating this result to formula Eq. (S25), applied to an $N-1$-level lattice, yields

$$\frac{\sigma_N}{\sigma_a} = \sqrt{\frac{(N-1)\left(\bar{K}^{2/N(N-1)} - 1 \right)}{1 + (N-1)\sigma_a^2 \bar{K}^{-2/N}}} \quad [S30]$$

We show this behavior in Fig. S2b for the case $\bar{K} = 0.1$, $\sigma_a = 0.001$.

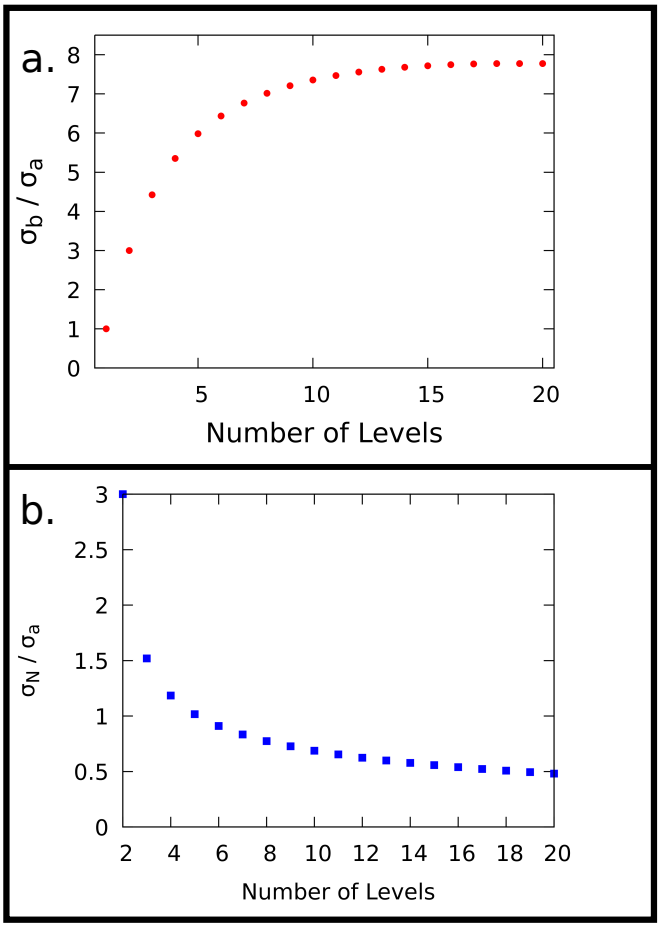

Fig. S2. a. The ratio of the standard deviation of bond portion on the 'new' level to the standard deviation of bond portion for the 'original' hierarchical levels at which the variation in stiffness of the new structure is equal to the variation in stiffness of the one level structure. The new level can always be assembled with a higher variation in bond portion. The effect saturates at a large number of levels, and allows for a striking amount of imprecision in assembly. **b**. The ratio of the standard deviation of the Nth level to that of the other $N-1$ levels, such that adding the Nth level does not increase the relative variation in stiffness.

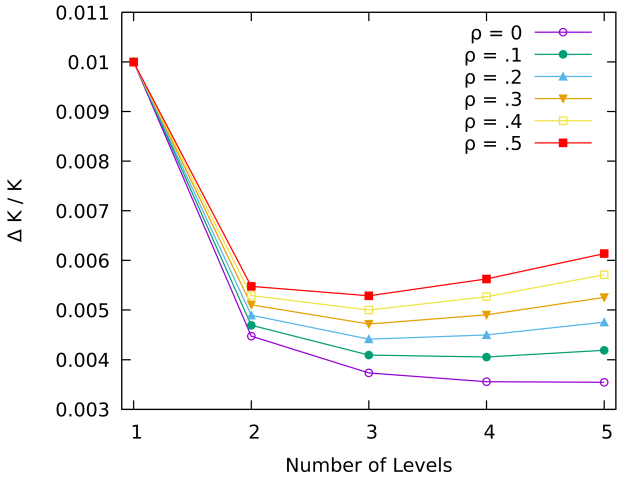

Fig. S3. The relative error is plotted against the number of hierarchical levels for cases in which the product of excess bond portions is 0.1, and the diagonal elements of the covariance matrix are 10^{-6}, and ρ is varied from 0.0 to 0.5.

We also consider the case in which the errors in bond portion on different structural levels are identically distributed, but correlated. For a network with N structural levels, we consider an $N \times N$ covariance matrix $\mathbf{\Sigma}$ which takes the form

$$\Sigma_{i,j} = \begin{cases} \sigma^2, & i = j \\ \rho\sigma^2, & i \neq j \end{cases} \qquad [S31]$$

It can be shown that

$$|\mathbf{\Sigma}| = \sigma^N \left[1 + (N-1)\,\rho\right](1-\rho)^{N-1}$$

In this case, the probability distribution function for a vector of bond portion errors $\vec{\delta}$ is given by

$$\mathcal{P}\left(\vec{\delta}\right) = \frac{1}{(2\pi)^{N/2}\,|\mathbf{\Sigma}|} \exp\left[-\frac{1}{2}\vec{\delta}^T\,\mathbf{\Sigma}^{-1}\,\vec{\delta}\right] \qquad [S32]$$

To illustrate this point, we show a plot of relative error in stiffness vs. number of levels, with fixed σ and varying coupling strength ρ (Fig. S3). While large correlations between errors eventually cause $\frac{\Delta\bar{K}}{\bar{K}}$ to increase, we find that having more than one level of structure always decreases $\frac{\Delta\bar{K}}{\bar{K}}$.

In view of the results of these alternative investigations, we anticipate that protection against fluctuation in stiffness is a generic benefit of structural hierarchy, and does not depend sensitively on the precise details of the distribution of errors in assembly.

Varying the Large-Scale Bond Length and Width. Our model is liable to fail if the width of a large-scale bond is either too great or too small in comparison with the length of the bond. We first consider the case in which bonds are too wide; here, the network ceases to behave as a structure with two, disparate length scales, and appears more like a sheet with small perforations. This is because our hierarchical networks may be viewed as single-scale triangular lattices with holes removed from them. It may be shown that, for large bonds of length l and width w, each of these holes is an equilateral triangle with side length s given by

$$s = l - w\sqrt{3} \qquad [S33]$$

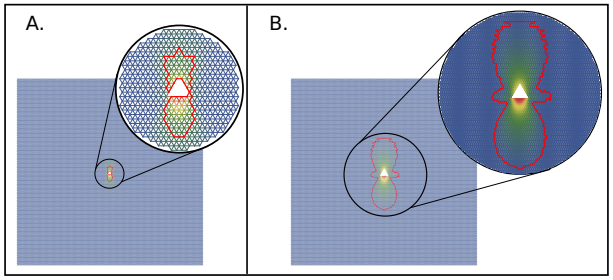

Fig. S4. The discrepancy between the displacement fields with and without a hole for **a**. a hole of length 5 and **b**. a hole of length 10. Here, we show annular sections taken from networks with overall dimensions of 200 by 200 bonds.

If each small bond has a length a, large bonds are n_l small bonds long, and large bonds have r rows of small-scale bonds, Eq. (S33) is equivalent to

$$s = a\left[n_l - \frac{3\,(r-1)}{2}\right] \qquad \text{[S34]}$$

We now seek the region over which the addition of a hole to an otherwise unbroken sheet has an appreciable affect upon the displacement field of a sheet under tensile strain, with the top and bottom pinned in the y direction, but free to relax in the x direction. We begin with a reference network, with overall dimensions of 200 by 200, in units of small-scale bond length, with no hole, and apply a strain of 0.05% in the vertical direction. We then apply this same strain to networks with holes in their centers, with hole side lengths varying from 2 to 15, in units of small-scale bond length. In each case, we subtract the displacement field at each point in the reference network from the displacement field in a network with a hole, omitting those points removed by the formation of the hole.

In seeking the region within which a hole has appreciably altered the mechanics of the network, we apply the following criterion: if the magnitude of the discrepancy between the displacement field in the presence of a hole and the displacement field in the absence of the hole is at least the applied strain times the small-scale bond length, the point lies within the "region of influence" of the hole. That is, if $\vec{u}_{ref}$ is the reference displacement field, $\vec{u}_{hole}$ is the displacement field with the hole, and ε is the applied strain, then

$$|\vec{u}_{hole} - \vec{u}_{ref}| \geq \varepsilon a \qquad \text{[S35]}$$

where εa is the approximate elongation of a small-scale bond in a perfectly affinely deformed network with overall strain ϵ. Below, we show two cases: a deformed network with a hole of length 5, and a deformed network with a hole of length 10. Networks are colored to show the magnitude of the discrepancy between the displacement fields in the networks with and without the hole, and the boundary of the holes region of influence is shown with a bold, red stroke (Fig S4).

While the region of influence is modest for small hole size, the region of influence for large hole size is considerable. To set a standard for widest acceptable width, given a certain length, we calculate the maximum difference from the center of the hole to a point on the perimeter of the center of influence. We denote this distance by r_{max}. For the lattices we consider, the center-to-center separation of holes of length l, separated by a single large-scale bond, is $\frac{l}{\sqrt{3}}$. Therefore, the size of the region

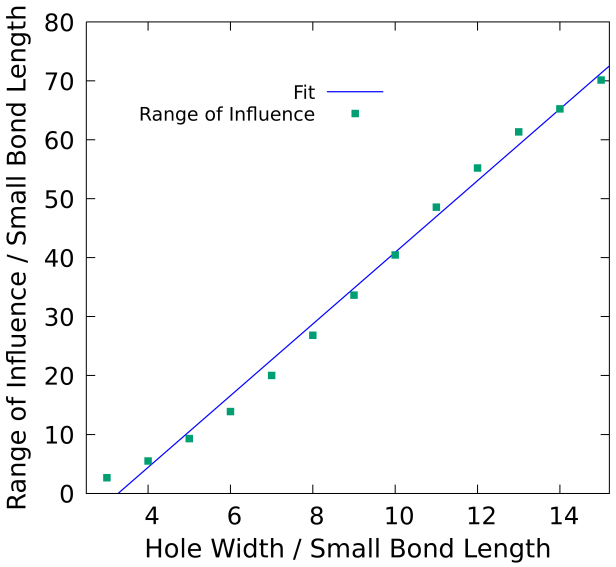

Fig. S5. Region of influence size vs. the width of a hole placed in the center of the network.

of influence of a hole should be comparable to or greater than this length. We find that, for holes with side length s of 3 or greater, in units of small-scale bond length, r_{max} is roughly linear with hole size:

$$r_{max} \approx 5.6s - 15 \qquad \text{[S36]}$$

with $r^2 = 0.99$. This demands

$$\frac{l}{\sqrt{3}} \leq 5.6s - 15 \qquad \text{[S37]}$$

$$\frac{l}{\sqrt{3}} \leq 5.6\left(l - w\sqrt{3}\right) - 15 \qquad \text{[S38]}$$

or

$$l - 2w \gtrsim 3 \qquad \text{[S39]}$$

We show data in Fig S5. While we have chosen to study networks in which springs have a stiffness of one, in units of one-dimensional stretching modulus over small-scale bond length, the equilibrium configurations of networks in their strained states would remain equilibrium configurations for the same loading conditions if the stiffness of each spring were re-scaled by the same multiplicative factor. We therefore find that the foregoing considerations should be of general applicability.

On the other hand, a bond may also be too narrow. Our analysis is sure to fail if large-scale bonds are only one or two bonds wide. In this case, the connectivity at the small scale is less than or equal to 4, and so the removal of even a small fraction of small-scale bonds will immediately destabilize the network. Further, overly narrow large-scale bonds will be highly susceptible to bending, which invalidates our picture of a primarily stretching-stabilized structure. In general, a thin rod of length l, with Young's modulus E, becomes unstable to buckling along direction $\hat{x}$ under a load

$$F_{cr} \propto \frac{EI}{l^2} \qquad \text{[S40]}$$

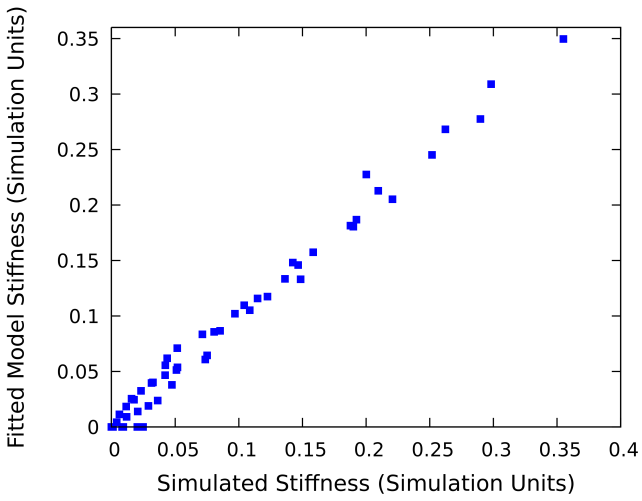

Fig. S6. Model vs. simulated stiffness values for a two-level network with large-scale bonds that are 5 rows wide and 20 small-scale bonds long.

with

$$I = \int x^2 dA \qquad [S41]$$

where the integral is over the cross section of the rod. (1) In two dimensions, for a rod of width w,

$$I = \frac{w^3}{12} \qquad [S42]$$

The prefactor in Eq. (S40) depends upon the precise boundary conditions at the ends of the rod, and is of order unity. On the other hand, we note that the network will generically contract in the direction transverse to applied tensile strain. Given a Young's modulus E for a large scale bond, a strain ε will lead to a force

$$F = wE\varepsilon \qquad [S43]$$

In all, Eq. (S40), Eq. (S42), and Eq. (S43) suggest bending will become a concern when

$$\frac{Ew^3}{l^2} \sim wE\varepsilon \qquad [S44]$$

or

$$\frac{l}{w} \sim \varepsilon^{-1/2} \qquad [S45]$$

We have also conducted an additional investigation to confirm that our model captures simulation results when the length and width of large-scale bonds relative to small-scale bonds are varied. We produced two-level networks in which each large-scale bond had five rows of small-scale bonds, and a length of twenty small-scale bonds. As in previous simulations, we varied both the small and large-scale bond portions from 0.55 to 1.0 in increments of 0.05, and as before, we found that our model captured simulation results. A plot of model prediction vs. simulation result for the 100 cases considered is linear, with a slope of 0.995, and an r^2 value of 0.991. Data are shown in Fig. S6.

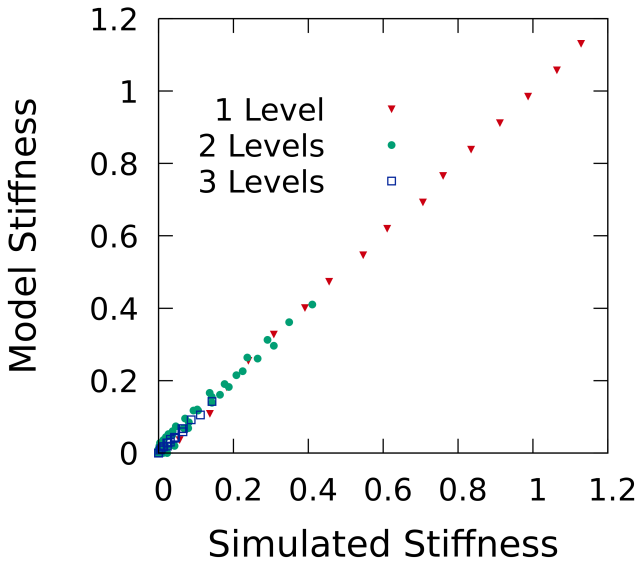

Fig. S7. Non-normalized stiffness values from the scaling model and simulation are compared. Despite the difference in maximum attainable stiffness, the same trend of strong agreement between model and simulation data is observed.

Non-normalized Comparison of Simulation and Model. In the main text, Fig. 2e shows a comparison of the model vs. the simulation stiffness values for networks with one-, two- and three-hierarchical levels, in which we normalized the stiffness value of a network with N levels by dividing that stiffness by the maximum attainable stiffness for an N level network. As before, stiffness units are the one-dimensional stretching modulus μ used in Eq. 5 in the main text, divided by the length of a small-scale bond. In Fig. S7, we show a modified version of Fig. 2e, in which stiffness values are not normalized. Notably, the maximum attainable stiffness decreases with the number of hierarchical levels, as the maximum density of small-scale units decreases significantly. Nevertheless, agreement between the simulation results and scaling model remains strong for each class of network considered, and within the range of stiffness values attainable, more hierarchical networks offer more reliability with less material used.

Assessing Stiffness Distributions for Differing Target Stiffness and Noise Values. In the main text, we discussed numerical trials in which we produced interpolating functions to predict the stiffnesses of hierarchical networks with specified bond portions on each scale, found combinations that would yield a desired, soft stiffness, and added zero-mean, Gaussian random noise to each bond portion over 50,000 trials. We found that the distribution of stiffness became progressively narrower with increasing levels of structural hierarchy. To assess the robustness of this effect, we have considered a range of stiffness values and noise standard deviations. Below, we show the results of the procedure described above in which stiffness values range from 0.02 to 0.05, in simulation units (for which the stretching stiffness of an individual bond is unity), and the standard deviation of noise added to bond portions varies from 0.005 to 0.001. We show heat maps for networks with one, two and three levels of structure, in which the mean absolute value of the difference between actual and target

stiffness, normalized by the target stiffness, is plotted vs. the standard deviation of noise and target stiffness (Fig. S8). In all cases we find that increasing the number of hierarchical levels, increasing the target stiffness, and decreasing the noise all decrease the error in stiffness.

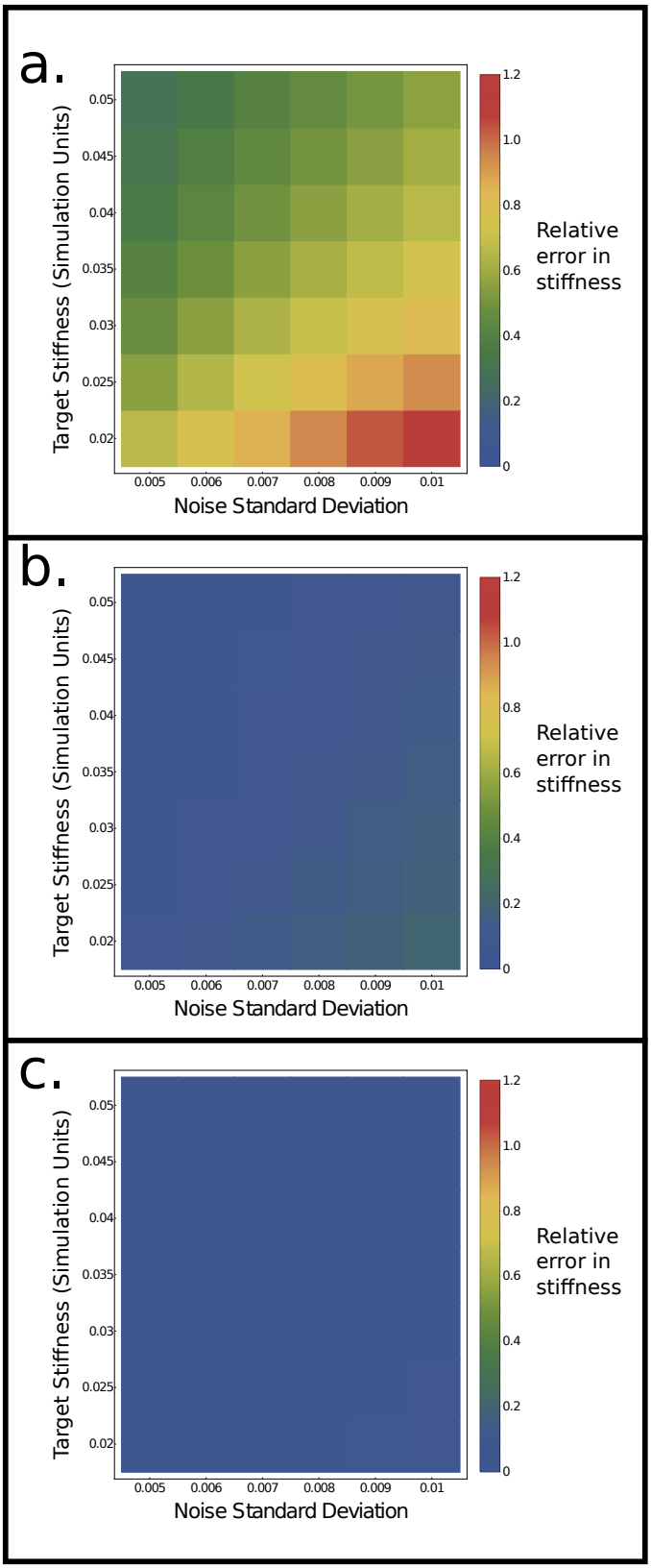

Fig. S8. Mean absolute value of deviation from target stiffness, normalized by target stiffness for **a** a network with one level of structure, **b** a network with two levels of structure, and **c** a network with three levels of structure.

1. Landau, L. D. and Lifshitz, E. M. *Theory of Elasticity, Second Edition* (Pergamon Press, Oxford), pp. 97-98.