Supporting Information

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SI Results and Discussion

1 Numerical and Analytical Results. To obtain analytical results for the relaxation of a single fiber, we performed calculations by using the mean field theory. There are several simplifying assumptions that are taken into account in these calculations. For instance, we assume that the concentration of enzyme particles is low enough to avoid correlations between particles that means that the particles do not interact with each other and the exclusion volume need not be included in the calculation. Also, we do not formally take into account the transition state in the model. The reason is that we focused more on the diffusion and concentration effects as well as on how tension on the fiber affects the relaxation of the force. This level of description of the local enzyme reaction is not necessary in the current model because we would not be able to extract such information from the experimental data. Consequently, the enzyme reaction is simply an on-off binding process. The chemical reaction step (peptide bond "hydrolysis") that includes interaction of the enzyme with the transition "state" is embedded within the probabilities p_{on} and p_{off}. The rate limiting step of the reaction is then governed by the lower value of these probabilities.

To obtain an expression for the number of particles leaving the fiber per unit of time $\langle\tau\rangle$, we assume that the number of bound particles n_B has reached the steady state as we show in Fig. S1. According to the results presented in the main text, p_{on} increases as the number of particle visits increases until it reaches the isotropic value of $1/3$. This behavior also affects n_B as can be seen from the cross-over region in all curves. As p_{off} increases, the cross-over region becomes shorter and n_B reaches the steady state earlier with a lower saturation value.

We can also calculate more explicitely the average waiting time $\langle\tau\rangle$ between two unbinding events. We first calculate the average number of particles leaving the spring $\langle n_L\rangle$ during one time step. Fig. S2 shows an array of 5 springs with $n_B = 3$ and $n_L = 2$ for the next time step. The probability P_{n_L} for two particles to leave the fiber at the next time step is related to p_{off} as follows:

$$P_{n_L} = 3p_{\mathrm{off}}^2(1 - p_{\mathrm{off}}).$$

From this particular case, we can generalize for any value of n_B and n_L as follows:

$$P_{n_L} = \frac{n_B!}{n_L!(n_B - n_L)!} p_{\mathrm{off}}^{n_L}(1 - p_{\mathrm{off}})^{(n_B - n_L)}.$$

By using the definition of the average value and the probability P_{n_L} above, we obtain

$$\langle n_L\rangle = \sum_{n_L=0}^{n_B} n_L P_{n_L} = n_B p_{\mathrm{off}}.$$

Thus, the rate of change of n_B is the difference between the number of particles that bind and the number of particles that leave the fiber per unit time as described in the main text.

2 Network. To mimic the geometry of a real elastin sheet, we build a two-dimensional network of fibers arranged randomly as described in the main text. Fig. S3 shows the network configurations before stretch (Fig. S3*A*) and after 20% uniaxial stretch in the positive x direction (Fig. S3*B*) at time 0. Notice that the network is homogeneous. After 200 time steps, the network becomes heterogeneous as shown by the colors (Fig. S3*C*). However, the displacements of the nodes remain in the neighborhood of their initial displacement (Fig. S4) that explains why the network shows an exponential decay of stiffness similar to that of individual fibers.

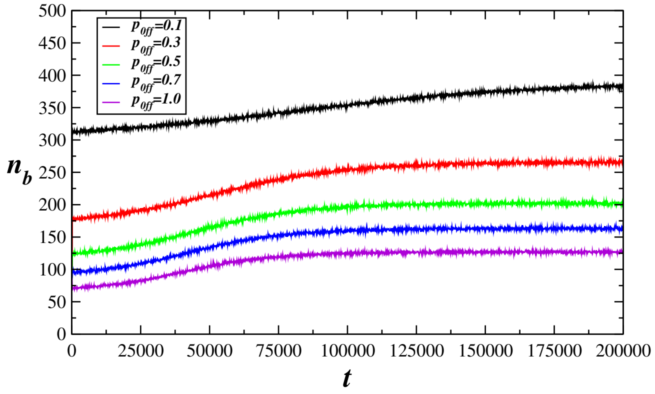

Fig. S1. The number of bound particles as a function of time for different values of p_{off}. The sum of the number of free and bound particles is constant and is given by $n_B + n_F = N_p = 512$.

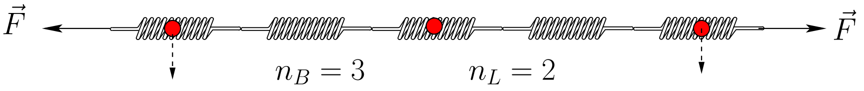

Fig. S2. An example of five springs with three bound particles. During the next time step, two of the particles leave the fiber.

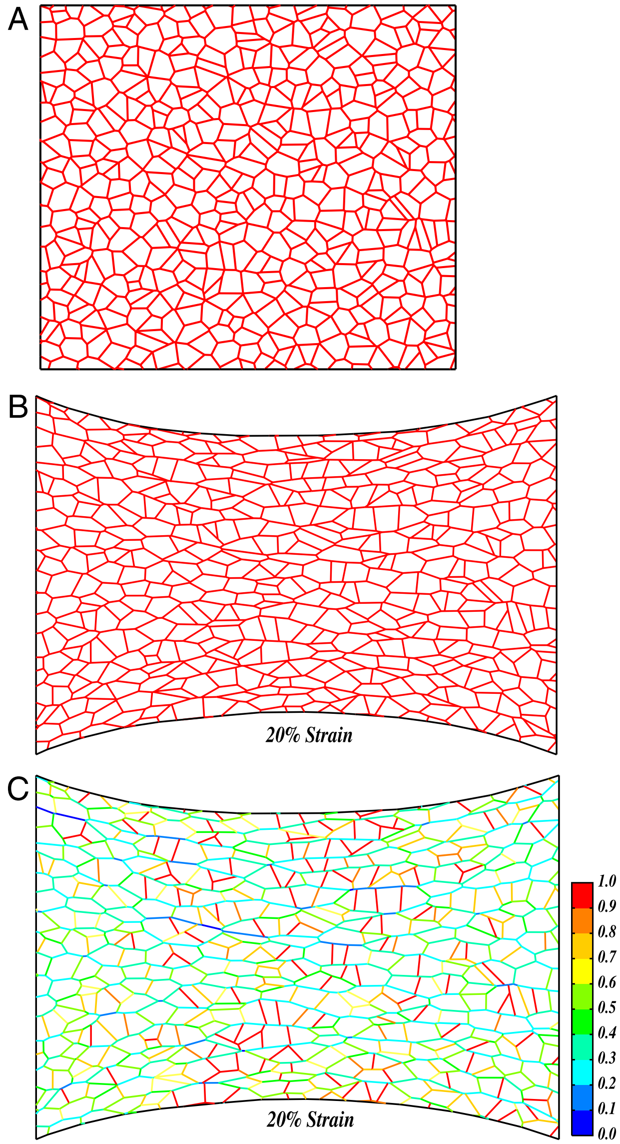

Fig. S3. In panel *A* the network is shown before stretch where the initial sizes are $L_x = L_y = 200$. The colors indicates the magnitude of the normalized spring constant $K/K_{\max}$ on each fiber (edge). All edges are red, which means that the spring constants are equal to unity. In panel *B*, the network is under tension but $t = 0$ and all spring constants are still equal to unity. In panel *C* $t = 200$ and the distribution of force on the network is heterogeneous, thereby generating different degrees of degradation on each spring (fiber).

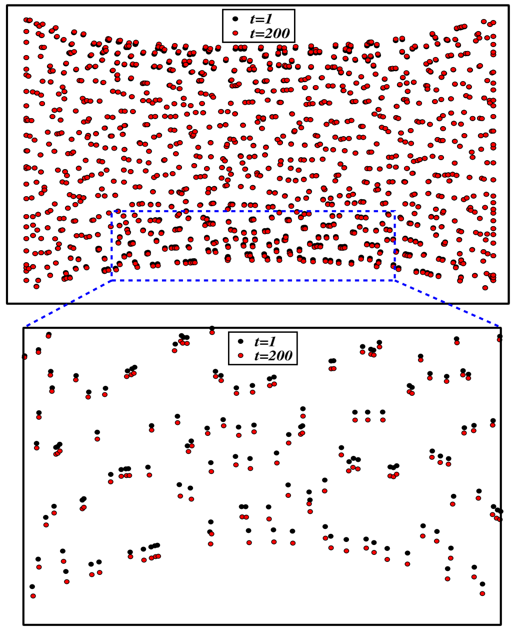

Fig. S4. The typical position of the network nodes at different times $t = 1$ and $t = 200$. In the *Inset* we can see the node displacements after the relaxation process takes place.