Electronic Supplementary Material (ESI) for Materials Horizons.

Stimuli-responsive Behavior of Composites Integrating Thermo-responsive Gels with Photo-responsive Fibers

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Supplementary Information

To emphasize the importance of the functionalized fibers in controlling the morphology of the sample, we determined the shape of stimuli-responsive gels that do not contain these fibers and are exposed to heat or light. Upon heating, the thermo-responsive LCST gel that is attached to the substrate shrinks symmetrically with respect to the front and back faces of the sample (i.e., does not bend, see Figs. S1a-b). This behavior is in contrast to the response of the sample containing fibers, where the composite bends towards the positive y-direction. Notably, when a *SP*-functionalized gel containing a *uniform* distribution of chromophores is illuminated with blue light (results not shown), the sample displays behavior similar to that in Fig.S1b.

When the gel without fibers is patterned with the spirobenzopyran chromophores in the same manner as the composite in Fig. 1a, it displays an asymmetric collapse (see Figs. S1c-d), and the morphology of the gel layer is similar to that in Fig. 2a. Namely, the entire sample is seen to bend in the negative y-direction. Here, however, the sample shrinks to a higher degree due to the absence of the fibers, which restricted the gel's collapse in the vertical direction in the example in Fig. 2a. (We note that in figure captions below, we use the words "fibers" and "posts" interchangeably.)

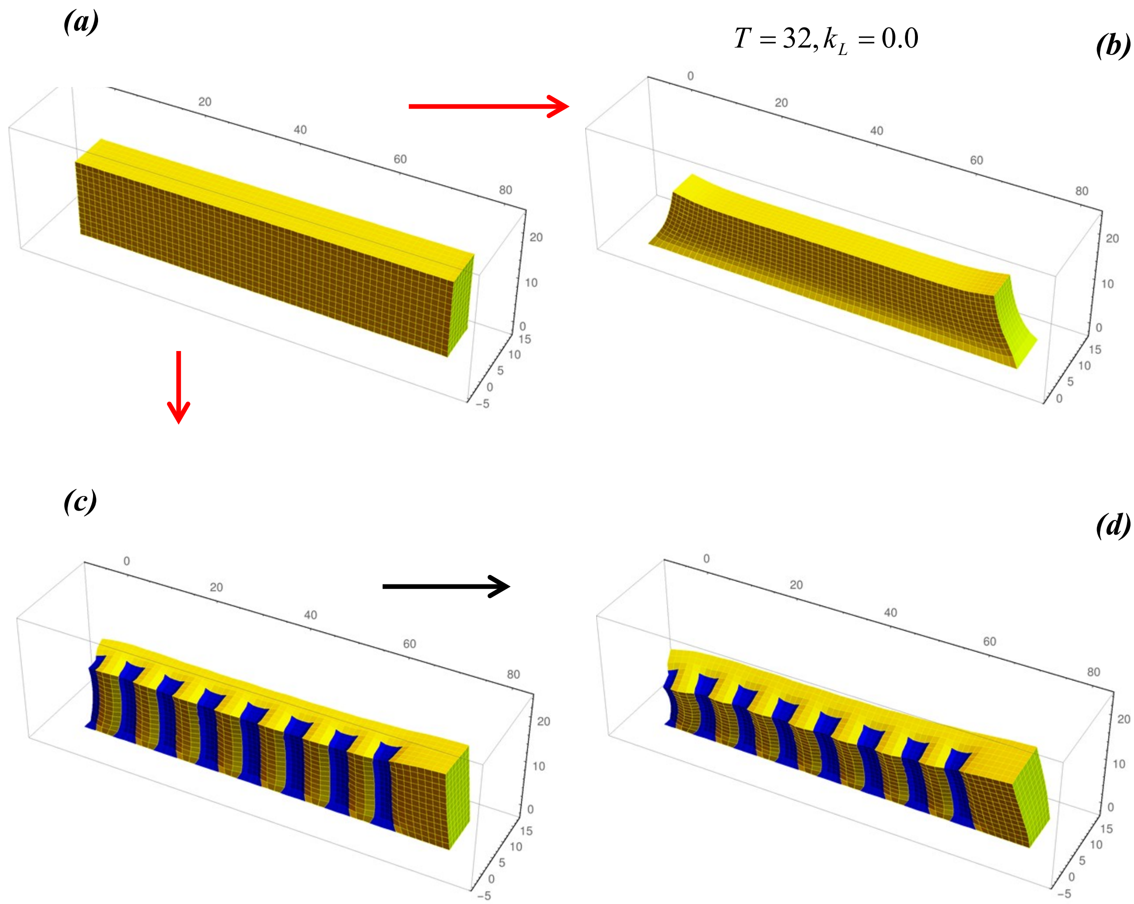

Figure S1. Response of a patterned gel without fibers to an increase in temperature **(a-b)** and illumination with light **(c-d)**. The *SP*-functionalized regions within the gel are located at exactly the same positions as in the composite in Fig. 2 of the main text. The image in **(b)** represents the steady-state shape of the heated gel in Fig. 2c in the absence of the fibers; here, T=32^{0}C. Similarly, the image in **(d)** represents the steady-state shape of the illuminated, patterned gel (with the patterning in Fig. 2a) in the absence of the fibers. The dimensionless simulation times are as follows: $t=0$ in (a), $t=3.2\cdot10^3$ in (b), $t=10^2$ in (c) and $t=3.2\cdot10^3$ in (d).

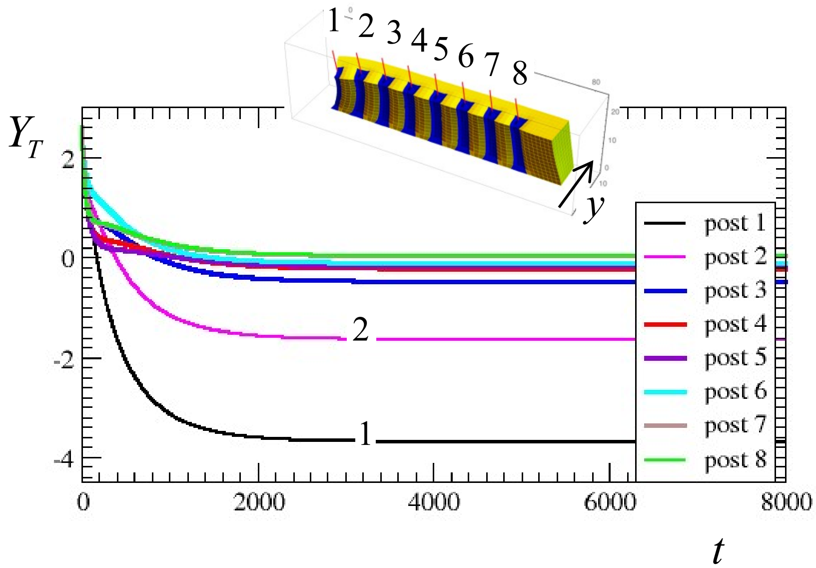

Figure S2. Time evolution of the *y*-coordinates of all the tips for the sample in Fig. 2a. We characterize the fibers' motion by the deflection of the middle fiber, which is the fourth post from the left (as marked in the inset). The dynamic motion of the posts follows similar behavior, except for posts 1 and 2. These terminal posts display a different behavior from the rest due to the distinct swelling of the gel at the free end and the corresponding bending of the left corner of the sample.

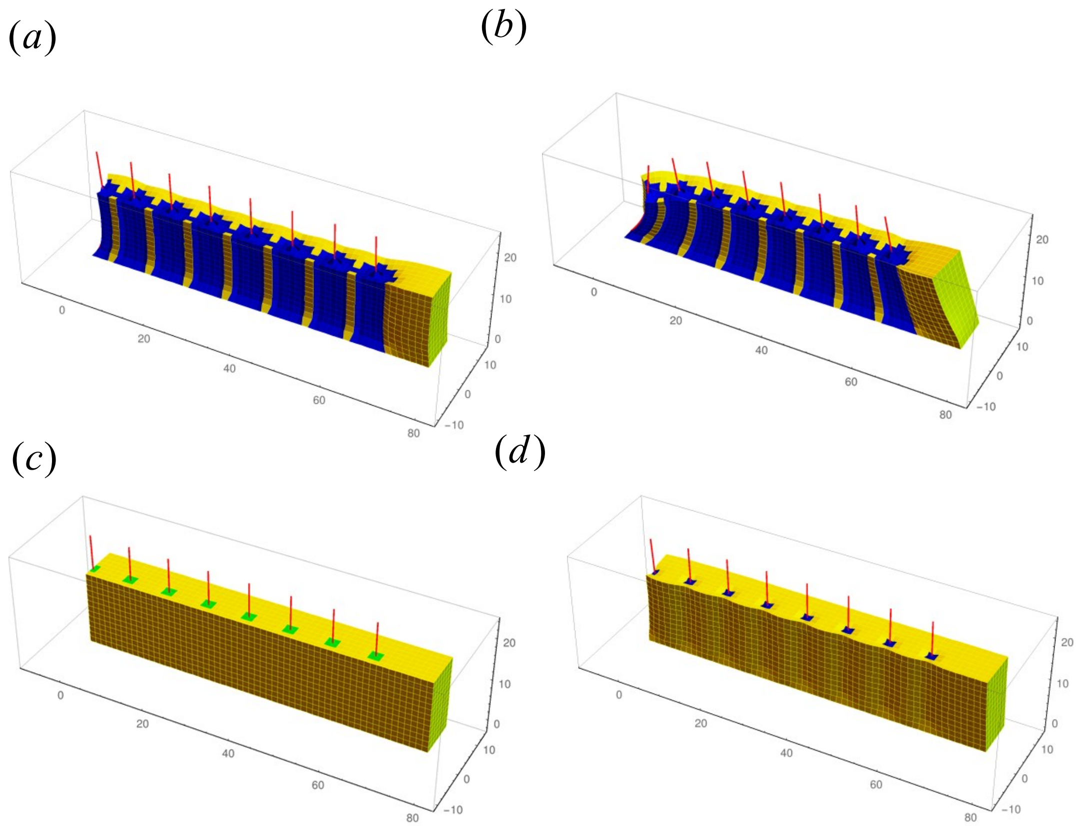

Figure S3. Dynamic behavior of the composites at different values of *R*, the radius of *SP* functionalization around the posts. Upper row: *R*=5, bottom row: *R*=1. Images on the left correspond to early times ($t = 20$ in **(a)** and **(c)**) and images on the right correspond to the steady-state shapes of the composites ($t = 6 \cdot 10^3$ in **(b)** and **(d)**).

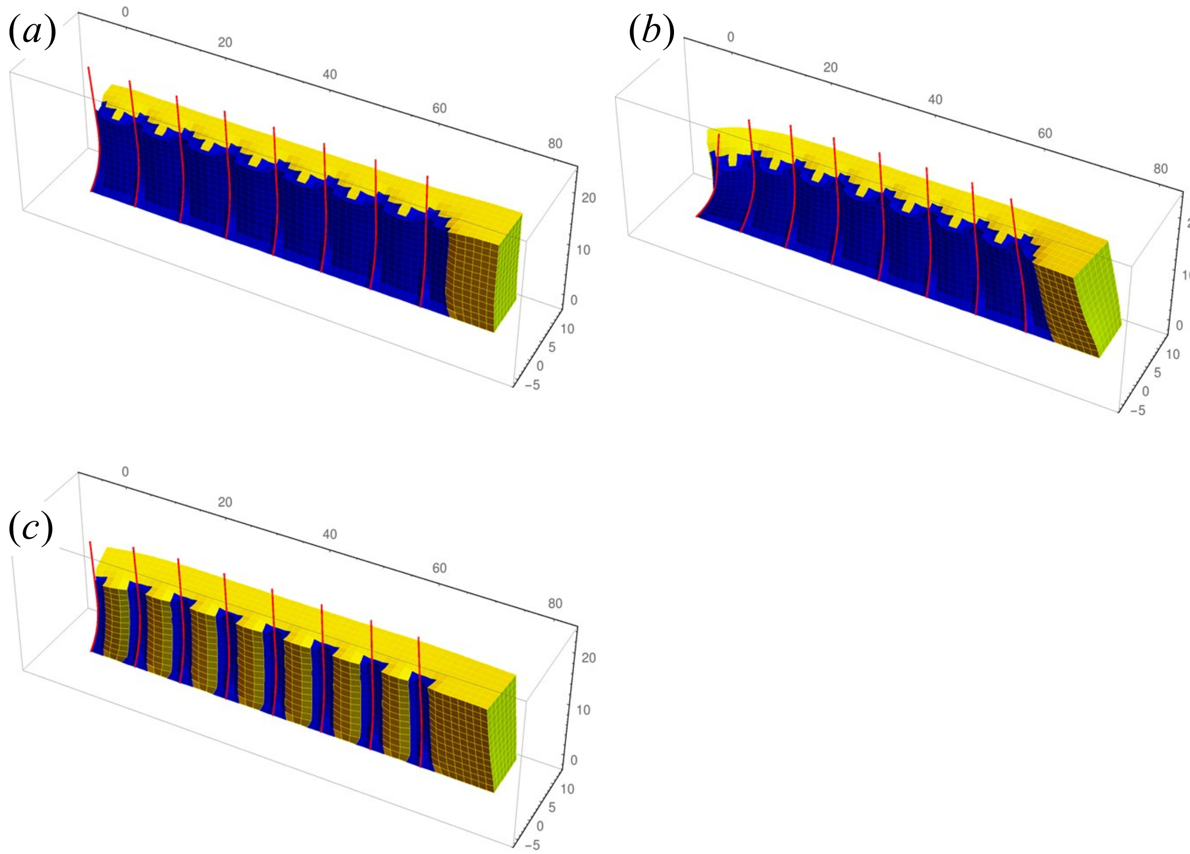

Figure S4. Dynamic behavior of composites with different values of *R*, radius of the *SP* functionalization around the posts, when these posts are placed directly at the front face of the gel, i.e., *p*=0. **(a-b)** Composite with *R*=5; here (a) corresponds to early times ($t=80$), and (b) to the steady-state ($t=6\cdot 10^3$). **(c)** Composite with *R*=3; image is taken at steady-state ($t=6\cdot 10^3$).

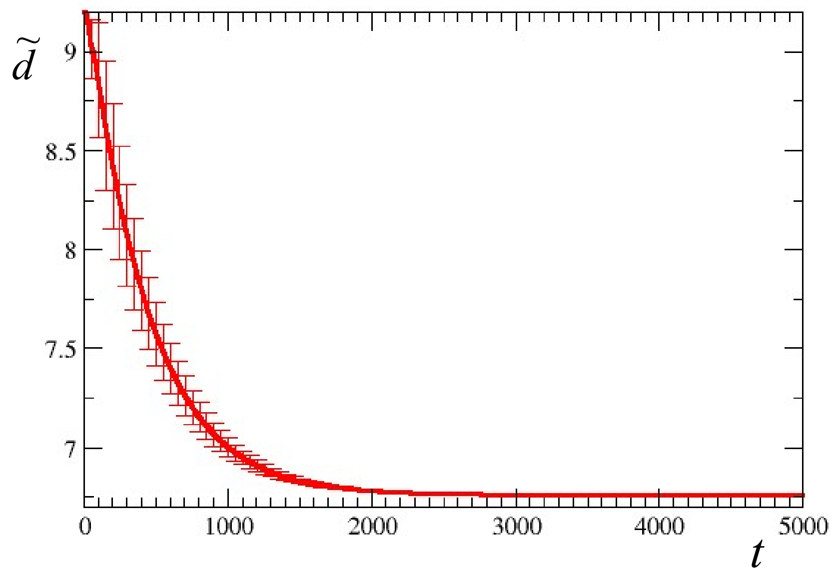

Figure S5. Evolution of the distance between the tips of the posts, $\widetilde{d}$, for the simulation in Fig. 6 of the main text. Here, we averaged over the distances between the tips of all posts except the two terminal posts. The posts are initially separated by $d = 7$ elements and move together with the elements; hence, the equilibrium distance between the posts at a given temperature can be calculated as $\widetilde{d} = d\lambda_{eq}$. The distance $\widetilde{d}$ is measured in dimensionless simulation units and is in excellent agreement with analytical estimates of the equilibrium degree of swelling of the gel provided in the *Model* section of the main text. Namely, we find that $\lambda_{eq} = 1.31$ at 20^{0}C and $\lambda_{eq} = 0.97$ at 32^{0}C, which results in the respective separations of 9.2 and 6.8 dimensionless simulation units.