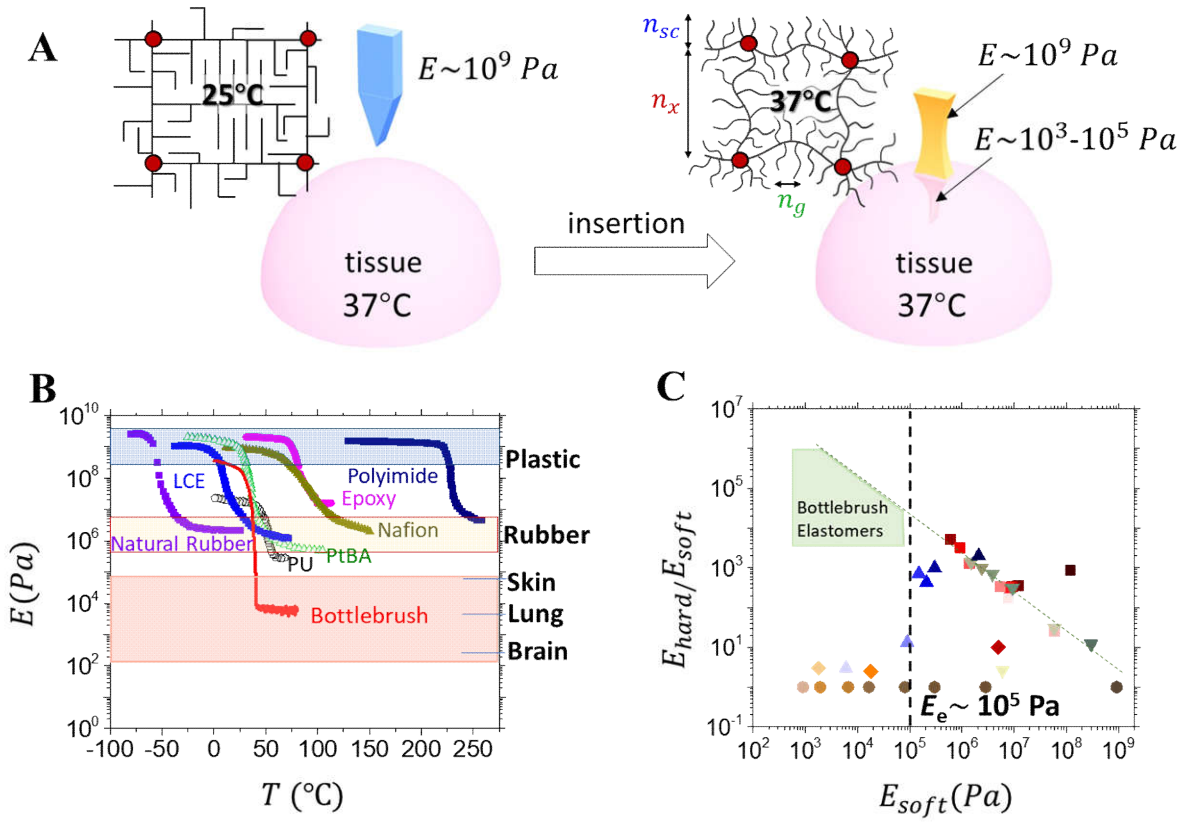

Figure 1: Hard-to-soft transition. A) Schematic representation of a tissue-adaptive implant. At a room temperature of 25°C, an implant is rigid, easily penetrating tissue followed by softening at a body temperature of 37°C to match the mechanics of the surrounding tissue (~10^2-10^5 Pa). B) Many polymeric materials undergo a sharp modulus decrease upon melting from a hard-state modulus of E_{hard}~1 GPa ("Plastic" bar) to a soft-state modulus of E_{soft}~1 MPa ("Rubber" bar) (Table S1). Their E_{soft} cannot go below ~10^5-10^6 Pa due to chain entanglements. In contrast, architecturally disentangled bottlebrush elastomers with crystallizable side chains allow a much greater modulus drop from the GPa to kPa level ("Tissue" bar). Furthermore, the brush-like architecture allows mimicking the mechanics of specific tissues such as skin, lung, and brain[6] by varying the architectural triplet [n_{sc}, n_g, n_x]: degrees of polymerization (DP) of the side chains (n_{sc}), of the spacer between neighboring side chains (n_g), and of the strand backbone (n_x).[7,8] C) Mapping conventional materials (including thermoplastics, gels, and tissues) by log-log plotting their modulus drop (E_{hard}/E_{soft}) against their E_{hard} (Table S2). Most thermoplastics (squares and downward triangles) have similar E_{hard}~1 GPa and, therefore, fall on the same line with -1 slope, but E_{soft} cannot cross the entanglement limit of E_e~10^5 Pa. Synthetic gels (upright triangles) and biological materials (circles and diamonds) can reach below this limit, but their softening transition is either marginal or nonexistent. With bottlebrush polymers (green polygon), the polymer trend can be extended below the 10^5 limit, yielding record high modulus drop (10^4-10^6 times) and tissue mimicking softness (10^3-10^5 Pa).

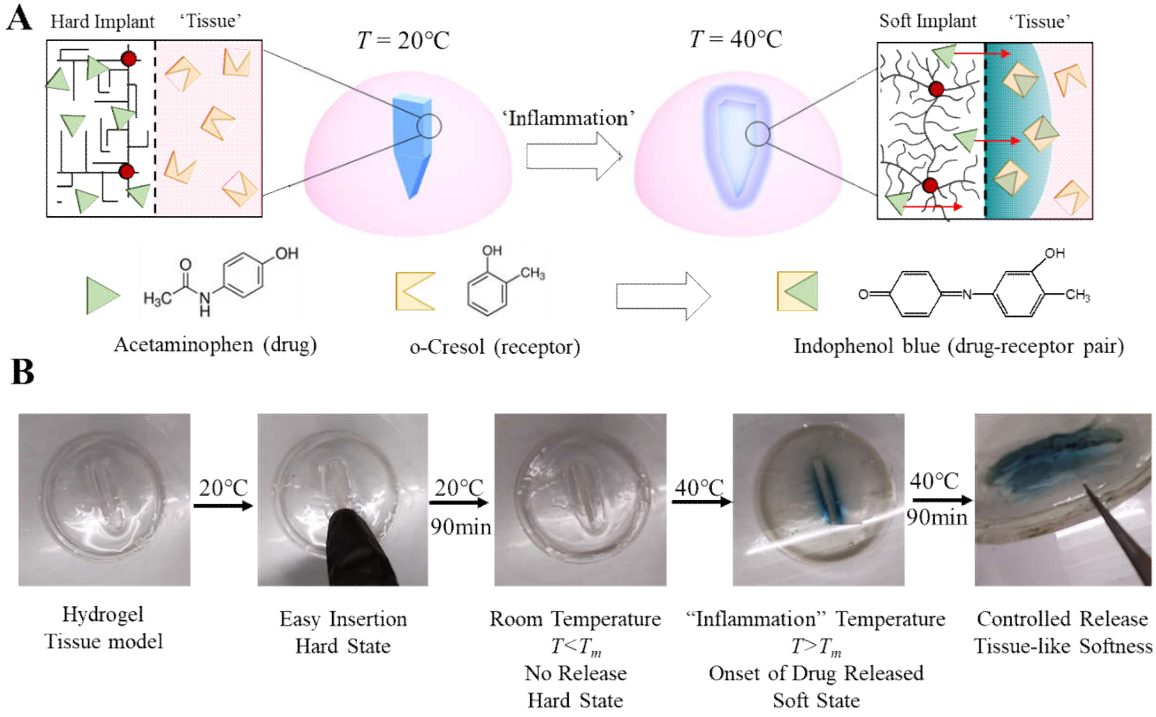

Figure 3: Imitated implant insertion and in-situ drug release. A) Schematics of an imitated drug release scenario, where increased temperature triggers the release of drug molecules followed by reaction with receptor molecules resulting in a visual indication. B) Polyacrylamide hydrogel (70 wt.% water) was used as a tissue model loaded with 5 wt.% water solution of o-Cresol molecules (acetaminophen indicators). An 'implant' was made out of a PVL bottlebrush elastomer ($n_{sc} = 10$, $n_{sc} = 50$) and loaded with acetaminophen (in 0.1 wt.% water solution). At room temperature (RT), a hard 'implant' is inserted into a hydrogel cavity followed by 90 mins incubation at RT with no visual indications. Upon increasing temperature to 40°C, an onset of blue coloration is observed around the 'implant'. During 90 min at 40°C, the coloration progressively enhances while the PVL implant remains clear and super-soft.

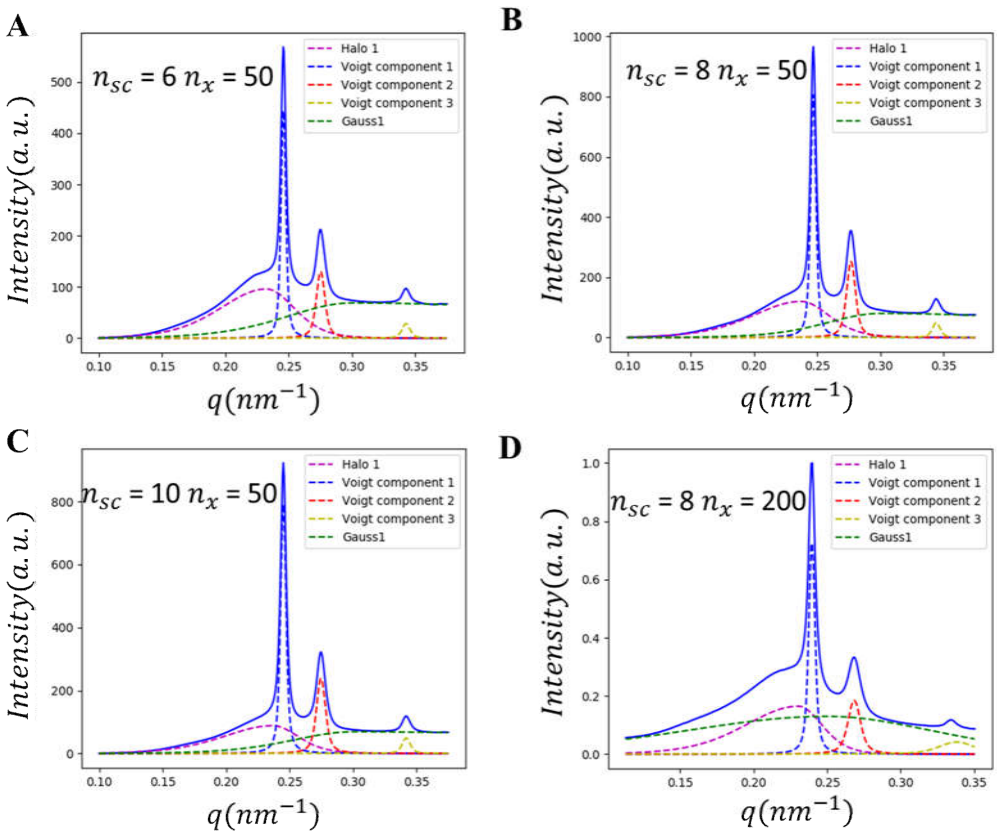

Figure S4: Crystallinity of PVL elastomers measured with SAXS measurements as shown in Table 1 for A) $n_{sc} = 6\ n_x = 50$ B) $n_{sc} = 8\ n_x = 50$ C) $n_{sc} = 10\ n_x = 50$ D) $n_{sc} = 8\ n_x = 200$ samples. The patterns are resolved into a polynomial background, an amorphous halo and (110), (200), (210) crystalline peaks. Crystallinity is calculated by the ratio of peak areas as $X_c = {A_1}/{(A_1 + A_2)}$, where A_1 is the sum of crystalline peaks and A_2 is the area of the amorphous halo. Compared to crystalline peaks, the intensity of amorphous halo decreases with increasing n_{sc}, and the similarity between $n_{sc} = 8\ n_x = 50$ and $n_{sc} = 8\ n_x = 200$ samples suggest n_x does not affect crystallinity.

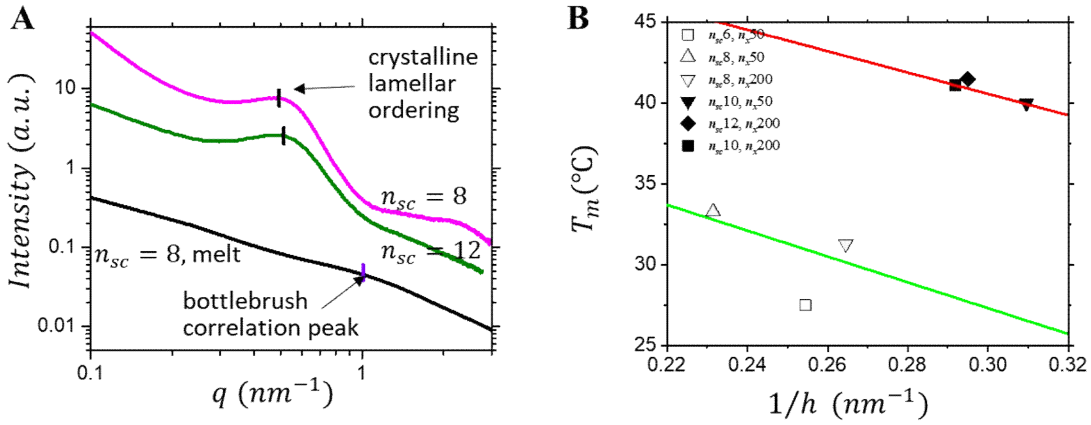

Figure S5: A) SAXS curves demonstrating effect of the side chain length on the structure of elastomers. Purple tick show bottlebrush backbone-to-backbone correlation peak position. Black ticks indicate interference peak in elastomers below T_m. B) Correlation between melting point (T_m) and crystal thickness (h) in the Gibbs-Thomson coordinates $T_m = T_m^0\left(1 - {2\gamma}/{(h\Delta H_0)}\right)$, where T_m^0 –

melting temperature of infinitely large crystal, γ - surface energy and ΔH_0 – melting enthalpy per unit volume. For the upper group, $T_m^0 = 333 \pm 28$ K, $k = 66 \pm 88$ K.nm, for lower group $T_m^0 = 324 \pm 21$ K, $k = 79 \pm 86$ K.nm.