Supplementary Data

The supplementary data aim to give an example of how one material property (G') of a degradable PEG hydrogel is affected by the addition of an adhesive ligand. The hydrogels for this experiment were 5% w/v precursor concentration and composed of 4-arm PEG-VS 10 kDa, PEG-diester-dithiol 3.4 kDa cross-linker [33], and 10 μM YIGSR ligand. The chemical structure of the cross-linker is presented in Figure S1. The degradation was achieved via hydrolysis of the ester bond present in the cross-linker.

The experiment was conducted as described in Materials and Methods, Section 2.5. Briefly, rheological measurements were performed with an AR 2000ex rheometer (TA Instruments) in parallel plate geometry, at 22°C, a frequency of 1-10 rad/s, and a constant 2% strain. The hydrogel samples were prepared to yield discs of 20-mm diameter and 1-mm thickness and were soaked in 10 mM phosphate buffer saline (PBS), pH 7.4 between measurements. The excess water from the hydrogel surface was carefully blotted before each measurement. G' at 1 rad/s was reported for each sample.

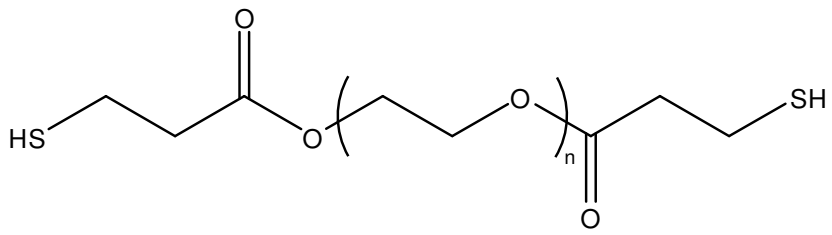

Figure S1: *Chemical structure of PEG-diester-dithiol where n = 77*

Figure S2 shows the change in G' upon PEG hydrogel degradation for hydrogels made with and without YIGSR ligand. As described in the body of the paper, addition of YIGSR at 10 μM concentration lead to an increase in G' (for the non-degradable hydrogels). The same

behavior was observed for the degradable PEG hydrogels. Moreover, the difference between the hydrogels made with or without ligand was not only retained but also increased over the course of the experiment. Initially, at 24 h, G' for hydrogels made with YIGSR ligand was 26% higher than G' for hydrogels made without ligand. The difference increased to 52% and 68% at 48 h and 72 h, respectively. The experiment was not carried further as the hydrogels became too weak to handle and degraded completely a few h after the last data point was taken.

The result from this experiment emphasized the fact that tethering hydrogels with ligands changed material properties such as stiffness (as represented indirectly through G') not only initially but even more so upon material degradation. These findings suggest the importance of assessing and controlling material properties upon addition of ligands, especially for applications wherein material degradation is advantageous (e.g., controlled release of drugs and therapeutic proteins).

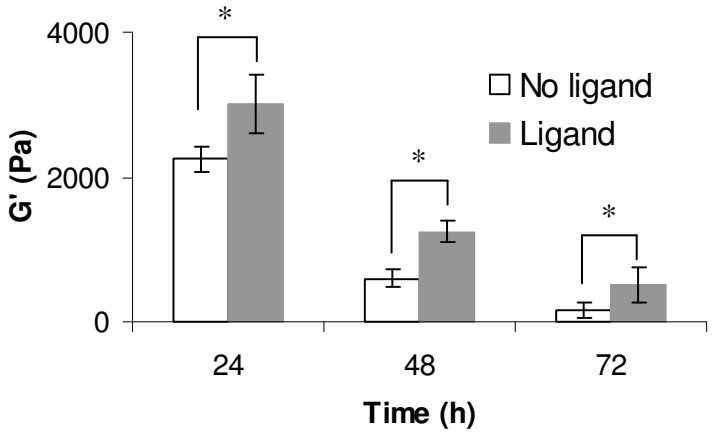

Figure 2: *Change in G' upon PEG hydrogel degradation for hydrogels made with or without YIGSR ligand. Asterisks designate significant differences.*