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Cytocompatible Tough Hydrogel Platform with Predictable Degradation

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Cytocompatible hydrogels with good mechanical properties and predictable degradation are highly desired for many biomedical applications including stem cell and therapeutics delivery for guided tissue repair. However, existing methods for fabricating tough hydrogels usually involve non-physiological conditions, such as toxic starting materials, catalysts, or crosslinking chemistry. Moreover, precisely controlling hydrogel degradation over a broad range in a predictable manner has been extremely challenging while empirical tuning of most degradable materials’ degradation profiles often resulting in undesired changes in other properties. To solve these problems, we recently developed a versatile hydrogel formulation that allows us to fabricate cytocompatible tough hydrogels under physiological conditions with predictable and widely tunable degradations. This platform was based on a well-defined hydrogel network formed by two pairs of four-armed poly(ethylene glycol) macromers terminated with bioorthogonal azide and dibenzocyclooctyl endgroups, respectively, via labile or stable linkages. The high-fidelity, catalyst-free bioorthogonal crosslinking reaction between these pairs of macromers enabled robust crosslinking in water, phosphate buffered saline and cell culture media to afford tough hydrogels capable of withstanding >90% compressive strain. The strategic placement of labile ester linkages near the crosslinking site within this superhydrophilic network, accomplished by facile adjustments of the ratio of the macromers used, enabled broad tuning of the hydrogel disintegration rates from 2 days to >250 days that precisely matched with the theoretical prediction based on a first-order linkage cleavage kinetics. This platform holds great potential for many biomedical applications that demands cytocompatibility, adequate mechanical integrity and precisely controlled temporal disintegration of the synthetic matrix.