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Spatiotemporal biomaterial functionalization via competitive supramolecular complexation of avidin and biotin analogs

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INTRODUCTION: Native tissues are characterized by a dynamic nature. Recapitulating such dynamicity in engineered tissues requires spatiotemporal control over their biochemical composition. Here, we pioneered supramolecular desthiobiotin/avidin complexation to enable dynamic functionalization of biomaterials. Desthiobiotin is a non-sulfur containing analog of biotin that also interacts with avidin, but with substantially lower binding affinity than biotin [1]. We hypothesized that combining biotinylated hydrogel with desthiobiotinylated bioactive molecules would enable the spatiotemporal functionalization of biomaterials via in situ desthiobiotin/biotin displacement, which is a novel, facile, and fully cytocompatible material modification strategy.

METHODS: Biotinylated dextran-based (Dex-TA) hydrogel constructs were functionalized with 1µM neutravidin (i.e., avidin analog), 1µM desthiobiotin-FITC (D-FITC) and 1µM biotin-atto565 (B-atto565), and subsequently analyzed using fluorescence recovery after photobleaching (FRAP) and confocal microscopy. Surface plasmon resonance imaging (SPRi) and reporter cells were used to analyze the reversible presentation and bioactivity of desthiobiotinylated BMP7 antibodies and BMP7 growth factors.

RESULTS & DISCUSSION: The reversible and sequential modification of the hydrogel constructs was demonstrated by displacing D-FITC with B-atto565. By tuning the concentration and incubation time of B-atto565, we reproducibly controlled its penetration depth into the hydrogel. An injection-molded bone-shaped 3D construct was spatially modified by controlling the thickness of the biotin-displaced layer. Moreover, competitive supramolecular complexation enabled the temporal presentation of desthiobiotinylated BMP7 antibodies and growth factors, as confirmed using SPRi and reporter cells.

CONCLUSIONS: We have successfully developed and characterized a novel spatiotemporal biomaterial modification strategy based on competitive supramolecular complexation.

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