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Hydrogels with Spatially and Temporally Controlled Properties to Control Cellular Interactions

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Stem cells (e.g., mesenchymal stem cells, MSCs) respond to many cues from their microenvironment, which may include chemical signals, mechanics, and topography. Importantly, these cues may be incorporated into scaffolding to control stem cell differentiation and optimize their ability to produce tissues in regenerative medicine. Despite the significant amount of work in this area, the materials have been primarily static and uniform. To this end, we have developed a sequential crosslinking process that relies on our ability to crosslinked functional biopolymers (e.g., methacrylated hyaluronic acid, HA) in two steps, namely a Michael-type addition reaction to partially consume reactive groups and then a light-initiated free-radical polymerization to further crosslink the material. With light exposure during the second step comes control over the material in space (via masks and lasers) and time (via intermittent light exposure). We are applying this technique for numerous applications. For example, when the HA hydrogels are crosslinked with MMP degradable peptides with thiol termini during the first step, a material that can be degraded by cells is obtained. However, cell-mediated degradation is obstructed with the introduction of kinetic chains during the second step, leading to spatially controlled cell degradability. Due to the influence of cellular spreading on MSC differentiation, we have controlled cell fates by controlling their spread ability, for instance towards osteoblasts in spread areas and adipocytes when cell remained rounded. We are also using the process of stiffening with time to investigate mechanically induced differentiation, particularly in materials with evolving mechanics. Overall, these advanced HA hydrogels provide us the opportunity to investigate diverse and controlled material properties on MSC interactions.