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The Ion-Specific, Non-Equilibrium Structural Behavior of DNA Hydrogels.

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— The highly tunable, sequence-dependent hybridization of DNA has enabled construction of DNA hydrogels with applications ranging from drug delivery to responsive materials. Though many have examined the structural characteristics of DNA hydrogels at equilibrium, relatively little is known about their non-equilibrium behavior, apart from their degradation rates when delivering molecular payloads. Here, we examine the effect of changing salt concentration on the dynamic formation, ageing, and degradation of DNA hydrogels comprised of branched DNA nanostars with palindromic overhangs. First, we observe that hydrogel phase is sensitive to the presence of a single unpaired base on the overhang, resulting in either a percolated network or a liquid-liquid phase separated state at high salt concentrations. Particular to the percolated network, we can induce the system to either contract or relax by changing the salt concentration. Decreasing monovalent NaCl induces the network to irreversibly contract whereas decreasing divalent $MgCl_2$ induces the network to reversibly expand; this behavior runs counter to what is expected solely from electrostatic screening. We qualitatively understand these results by assuming that the monovalent salt modulates the dynamic hybridization between nanostar binding partners, whereas the divalent salt drives the dramatic/reversible induction of the 'stacked-X' conformation in the DNA nanostars.

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