

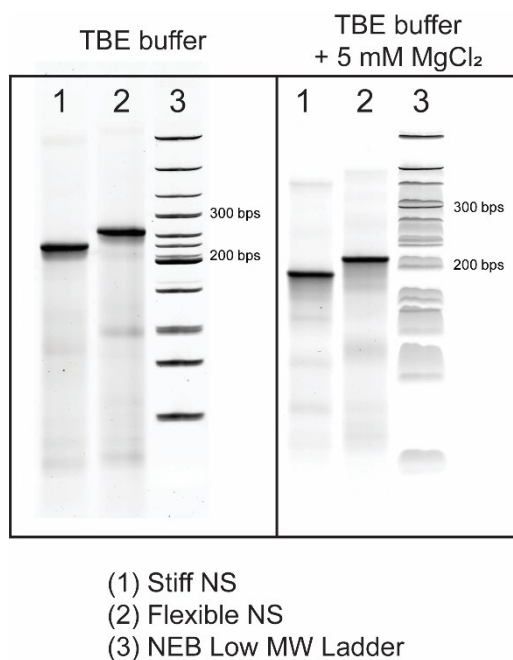
Electronic Supplementary Information (ESI)

Movie captions

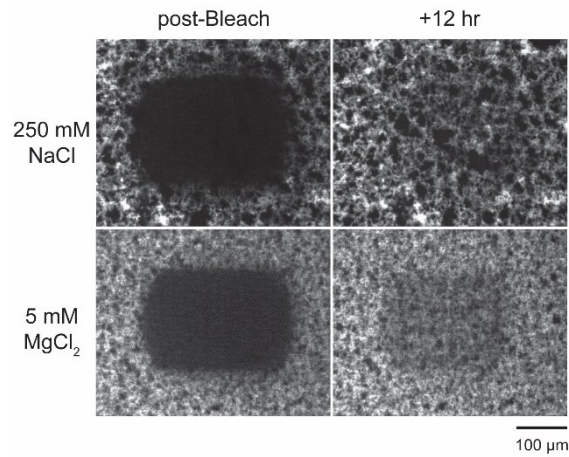
Movies S1 and S2: These videos show the time course of two NS systems as they respond to time-dependent changes in the salt concentration, particularly a quench from low to high salt concentration that stabilizes NS hybridization interactions. In both videos, the time label (bottom left) shows hours:minutes:seconds, while the salt concentration at each time is labeled in the top left. Movie S1 shows the maturation of a fully-flexible NS system into dense DNA droplets that sediment and coalesce. Movie S2 shows the percolation of networks of fully-stiff NS.

Movie S3: This video shows a permeable-membrane experiment on a fully-stiff NS system that begins in a contracted state (see Fig. 4a). Salt is switched from high to low, and the dissolution of the network is observed. Time and salt labels are as in Movies S1 and S2. We see that, just before dissolution, the hydrogel significantly expands.

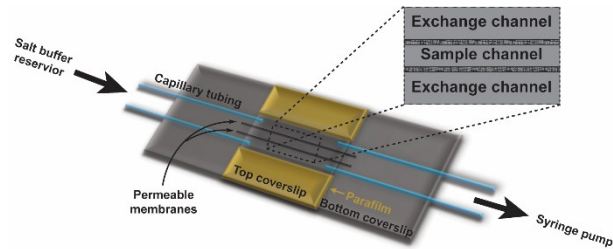
Supplementary figures and figure captions



Supplementary Figure S1: NS were examined using polyacrylamide gel electrophoresis (PAGE) to probe the efficiency of formation from the annealing procedure. NS lacking overhangs were used so as to remove the possibility of sticky-end interactions, thereby leaving two samples for analysis: blunt stiff NS and blunt flexible NS. Each sample exhibits a single dominant band of correctly formed 4-arm NS (left), matching published results;¹² as expected, flexible NS, with their additional bases between NS arms, migrate more slowly than stiff NS. Though dim bands are present both below and above the dominant band of each sample – corresponding to incomplete NS and dimers of incomplete NS, respectively – the vast majority of the DNA population is correctly formed NS. When PAGE is repeated in the presence of 5 mM MgCl₂, both stiff and flexible NS migrate further than in the absence of added salt, relative to the DNA ladder (right). This increased migration supports the notion that NS adopt a more compact “Stacked-X” conformation in the presence of MgCl₂.



Supplementary Figure S2: Even after 12 hours, the bleached spot is still visible in both 250 mM NaCl and 5 mM MgCl₂; this supports the slow dynamics of the gel/gel exchange process.



Supplementary Figure S3: Permeable membrane design: Within a flow cell constructed by melting parafilm between two glass coverslips, a central channel is separated from two buffer exchange channels by two PEG-diacrylate permeable membranes. Thin capillary tubes are inserted into the exchange channels and connected to a buffer reservoir and a syringe pump, enabling salt exchange to the system. NS samples within the central sample channel are unperturbed by flow in the exchange channels.