Supplementary Information

Precise and Selective Macroscopic Assembly of Dual Lock-and-Key Structured Hydrogel

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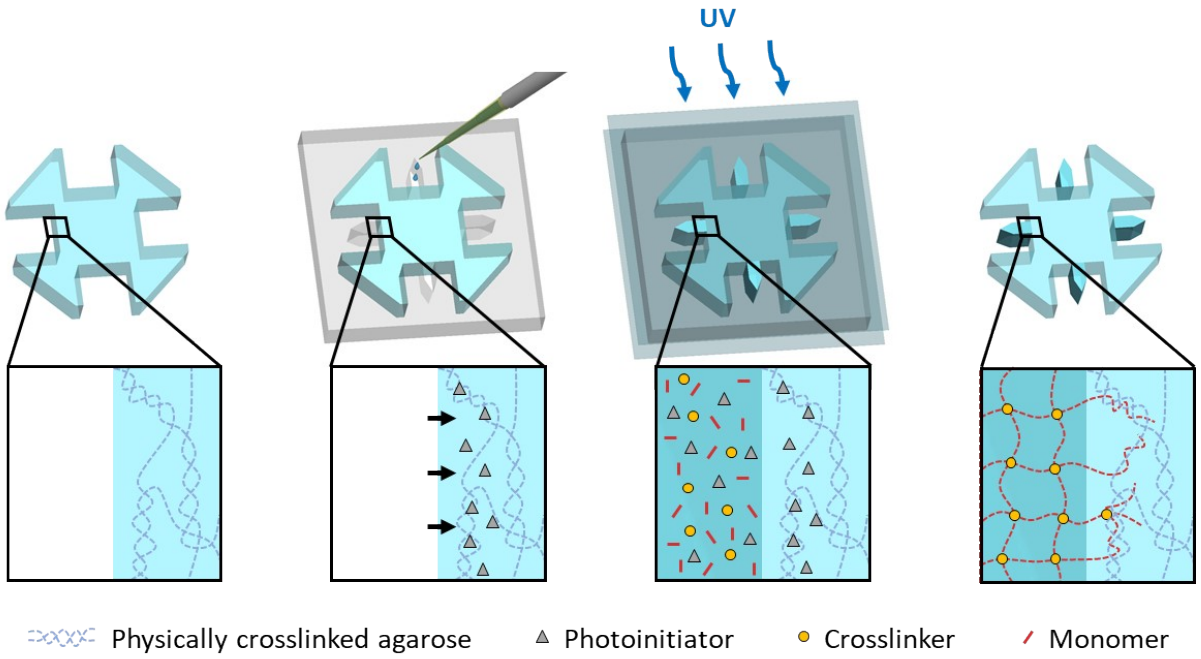

Fig. S1 Schematic illustration of the overall fabrication process for the chemically heterogeneous hydrogel building blocks.

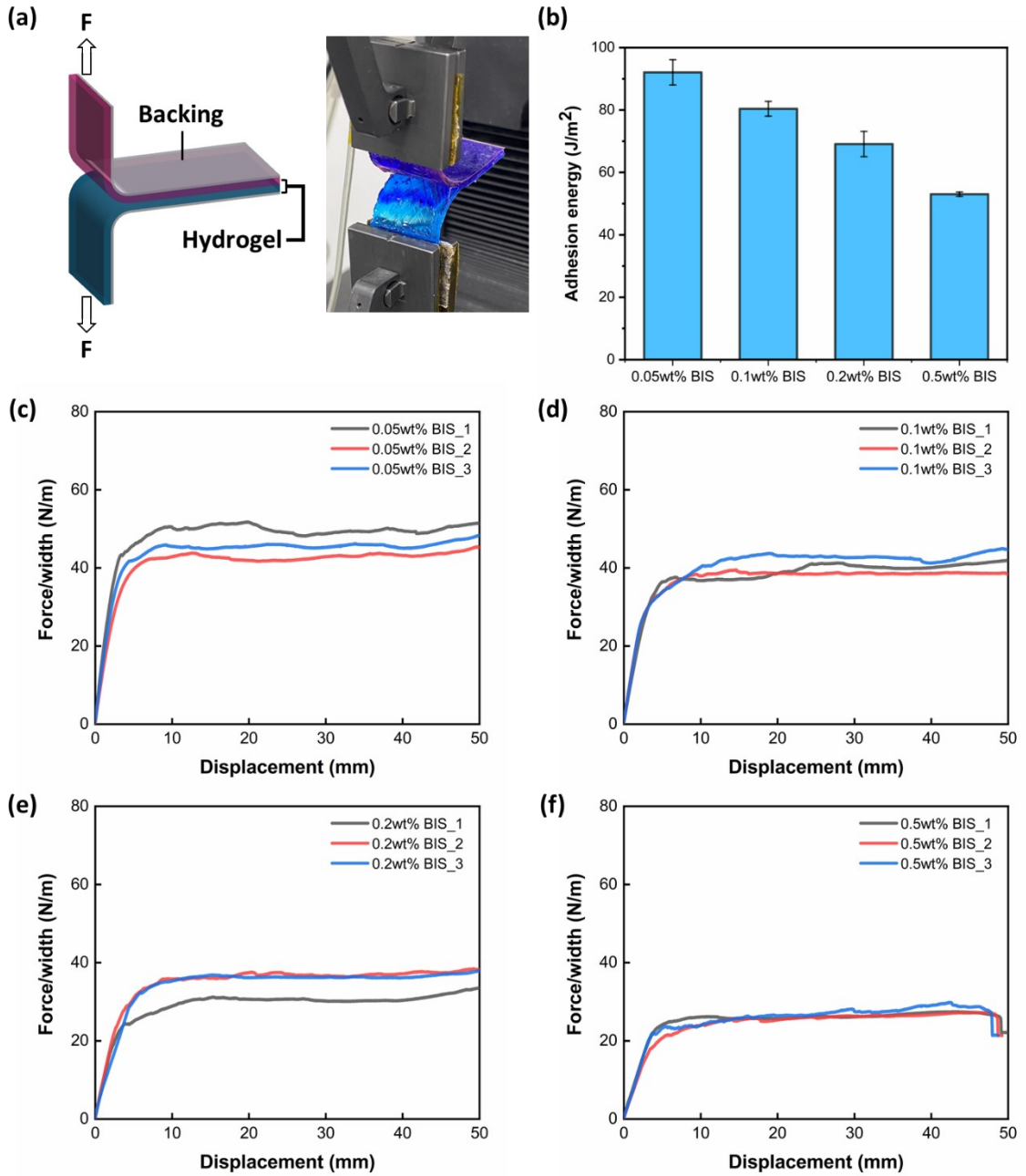

Fig. S2 (a) Schematic illustration and experimental image of a 180°-peeling test. (b) Adhesion energies between the oppositely charged hydrogels, varying the crosslinker concentrations. Representative force/width-displacement curved of the 180°-peeling test with crosslinker concentrations of (c) 0.05 wt%, (d) 0.1 wt%, (e) 0.2 wt%, and (f) 0.5 wt%.

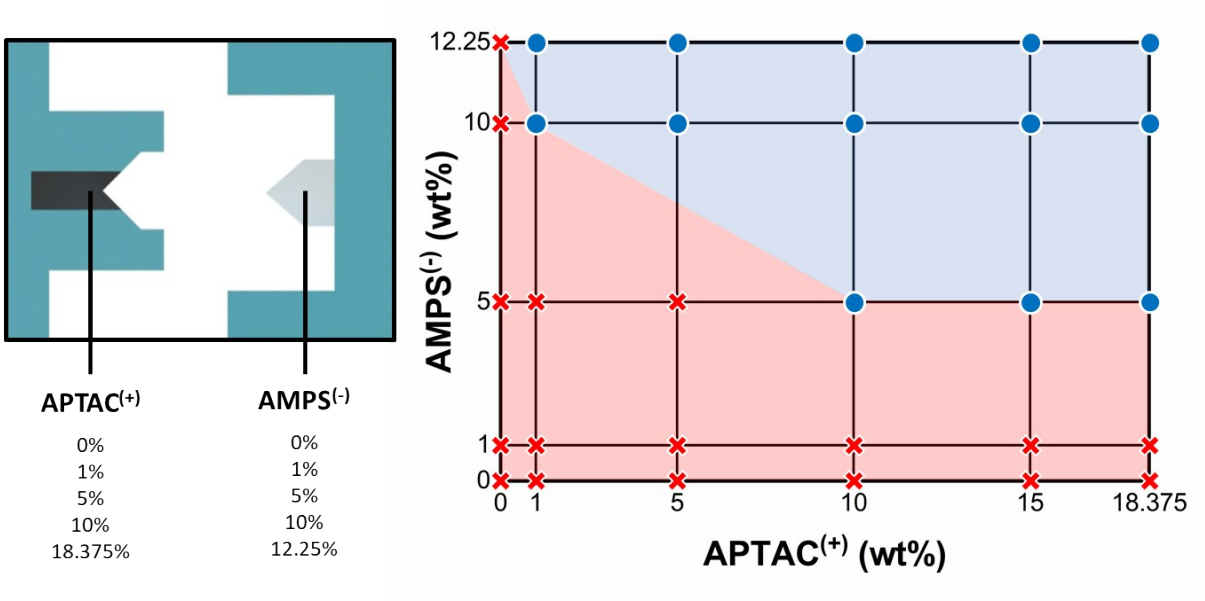

Fig. S3 Assembly stability test between charged hydrogels by varying the concentration of positively and negatively charged monomers.

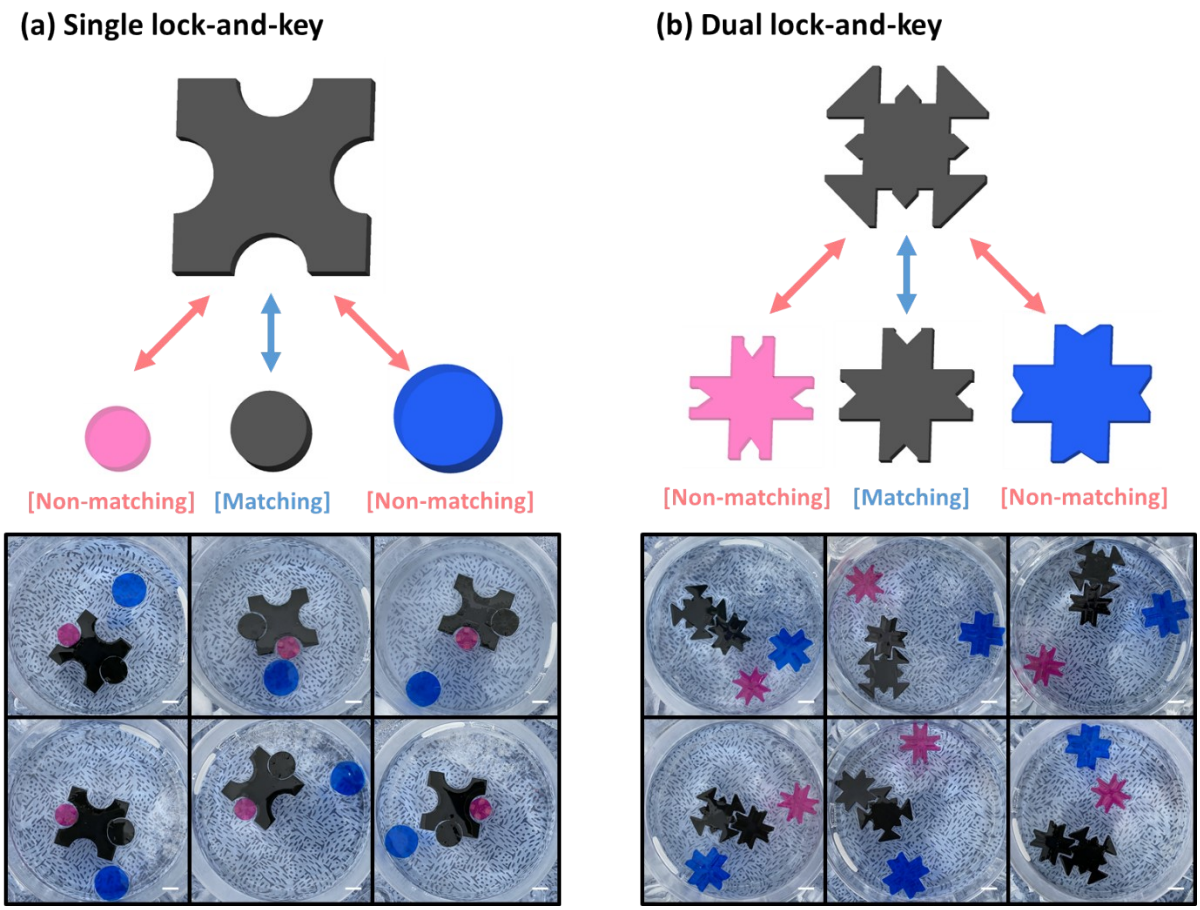

Fig. S4 Design schematics and experimental images displaying (a) assembly using the single lock-and-key structures, and (b) assembly using the dual lock-and-key structures. Scale bars: 1 cm.

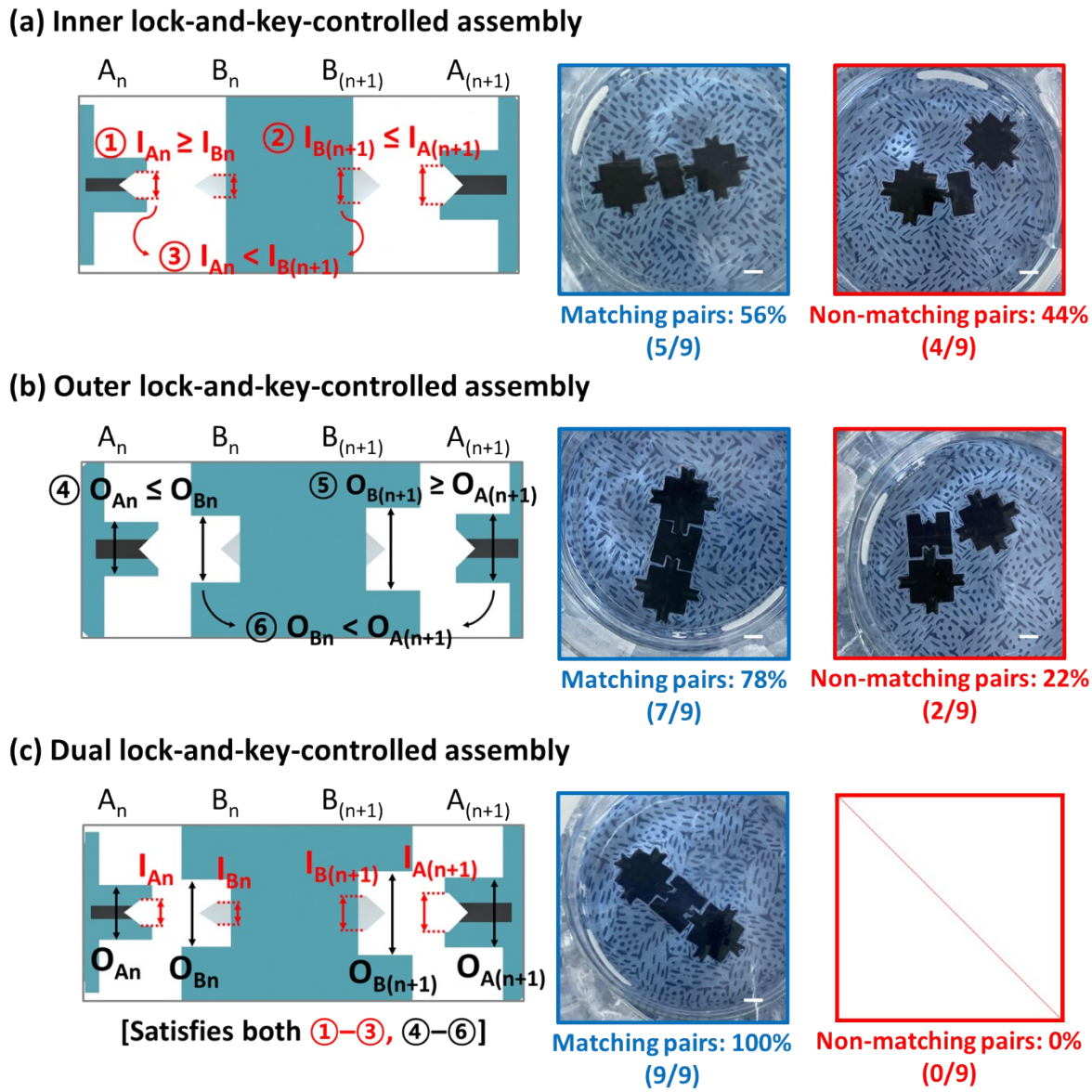

Fig. S5 Design schematics and assembly yields of the building blocks to assess the accuracy of the dual lock-and-key-assisted selective assembly when controlled by (c) the inner lock-and-key only, (d) the outer lock-and-key only, and (e) the dual lock-and-key. Scale bars: 1 cm.

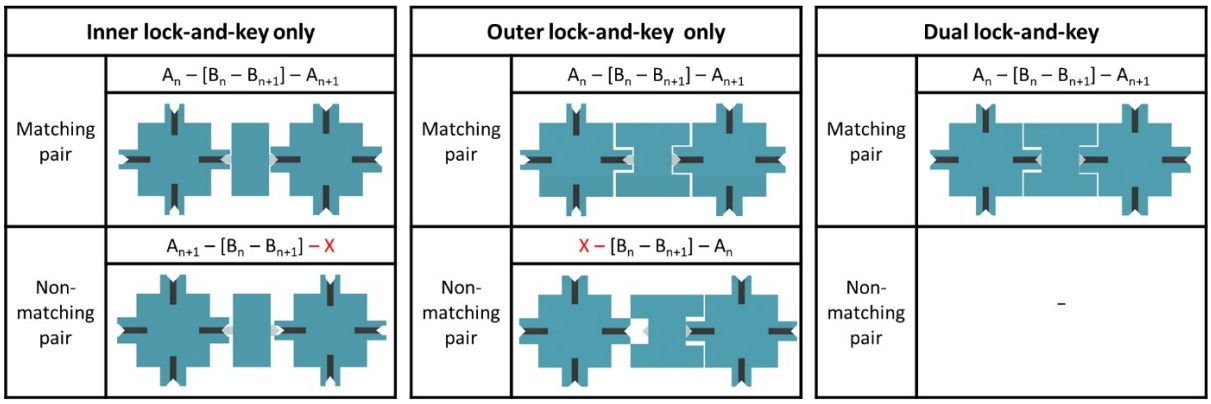

Fig. S6 Schematic illustrations of the possible matches and non-matches between the hydrogel building blocks, controlled by the two lock-and-keys depicted in Fig. 2c-e.

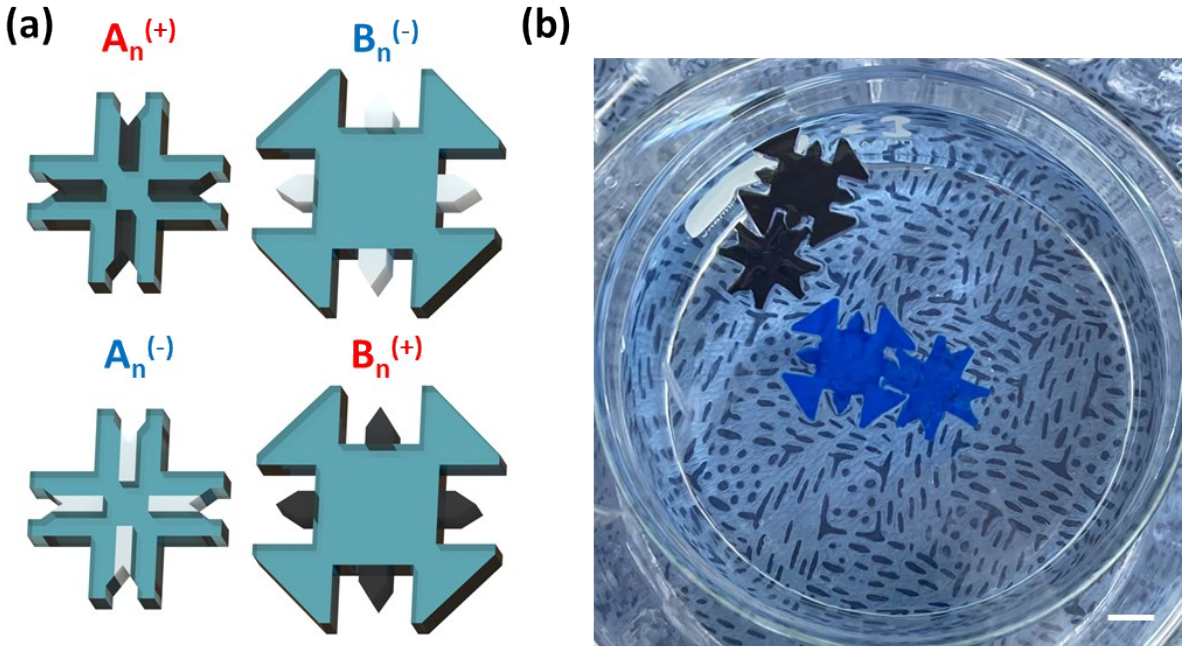

Fig. S7 (a) Schematic illustrations of two hydrogel building blocks with identical designs but different charge arrays. (b) Experimental image demonstrating selective assembly only between oppositely charged pairs. Scale bars: 1 cm.

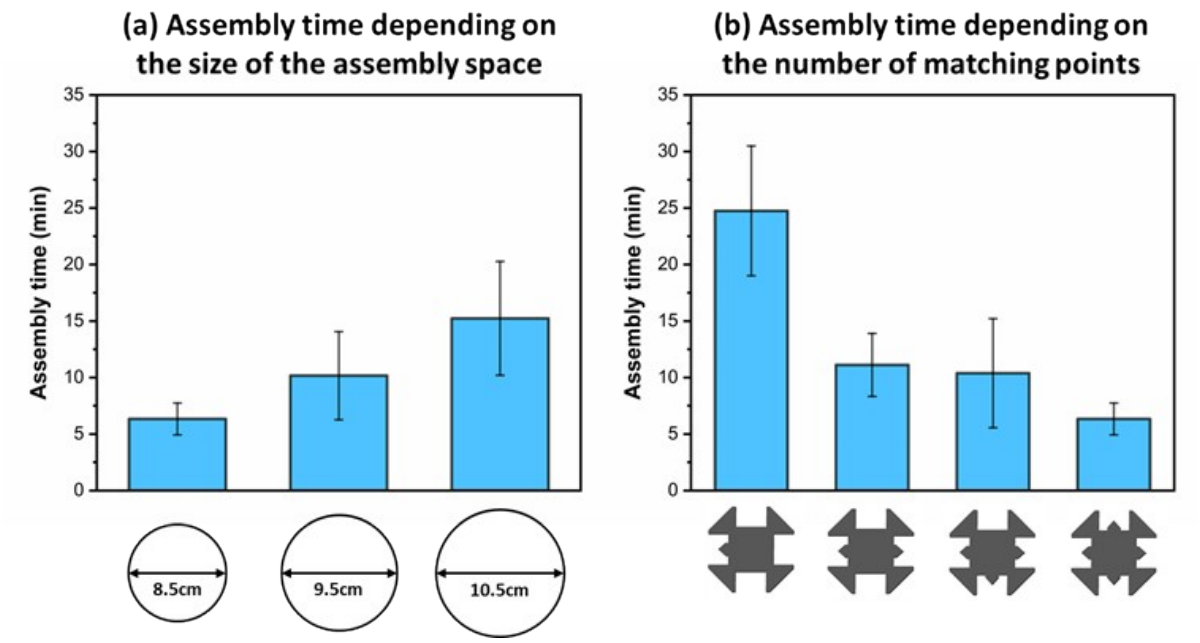

Fig. S8 Graphs depicting the time consumed for assembly using the dual lock-and-key structure depending on (a) the size of the assembly space while maintaining four matching points (n=5 for each case), and (b) the number of matching points in a single building block with an 8.5 cm dish size (n=5 for each case). The size of the hydrogel building blocks was 23.8 mm in width, while the size of the corresponding matching pair was 19.8 mm.

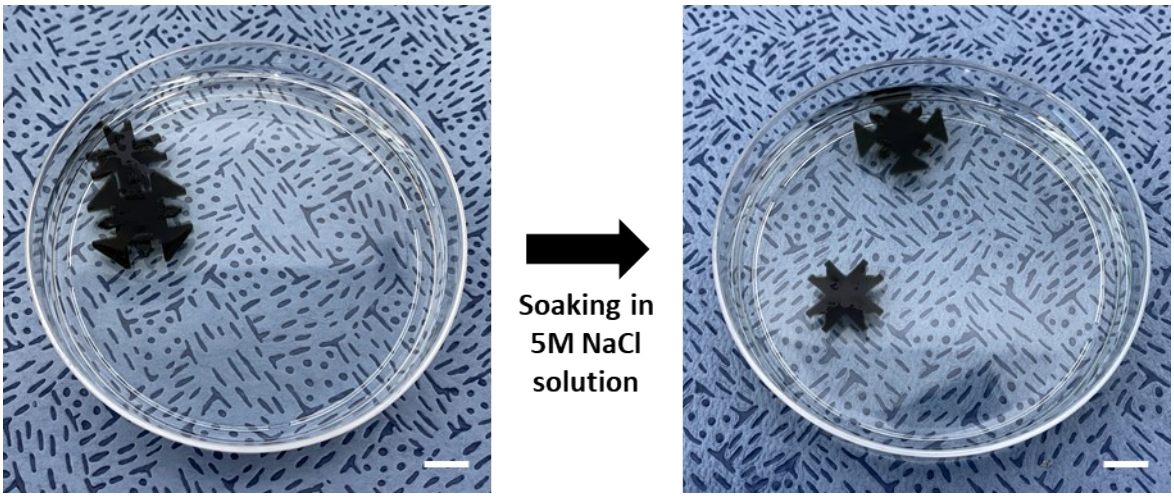

Fig. S9 Experimental images of demand detachment of the assembled hydrogel building blocks under salt conditions. Scale bars: 1 cm.

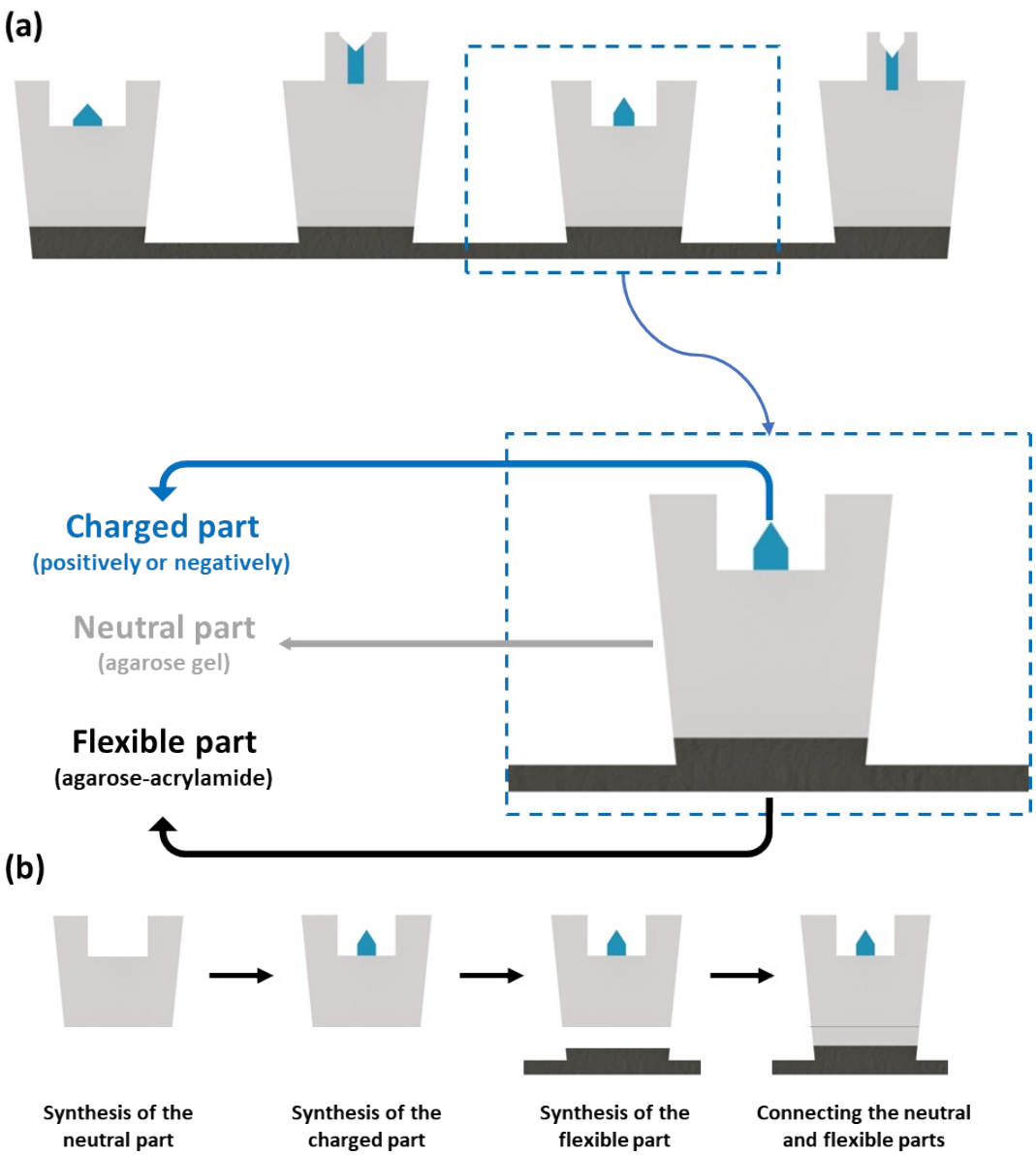

Fig. S10 (a) Schematic illustrations of DNA-mimicking double-stranded structures, consisting of three components: a charged part, a neutral part, and a flexible part. (b) Synthesis procedure for DNA-mimicking structures.

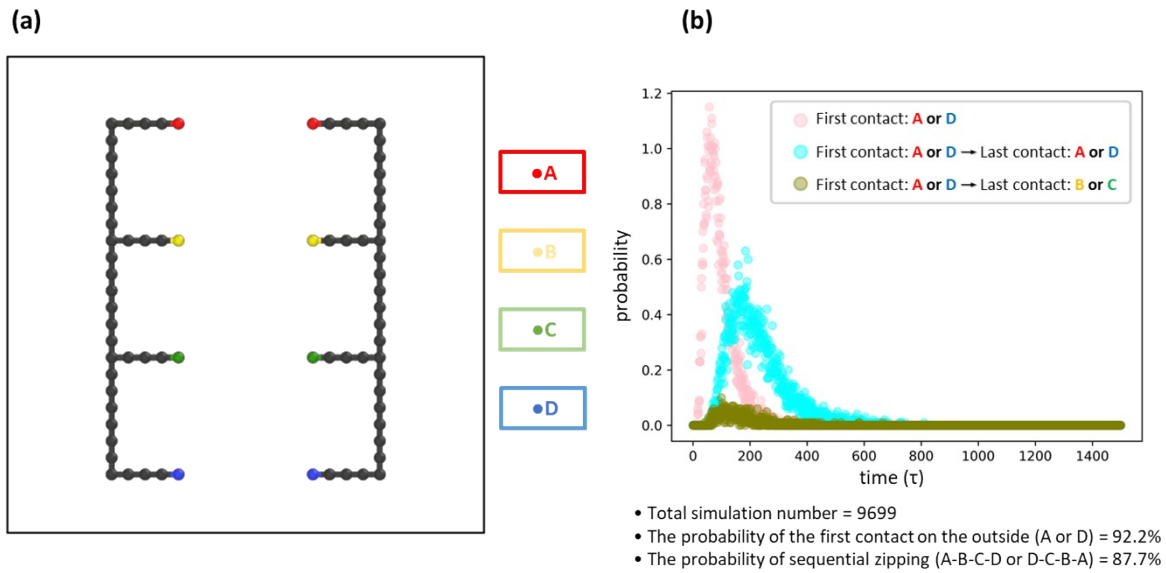

Fig. S11 (a) Designed model of a DNA-mimicking structure for molecular dynamics simulation. (b) Graph depicting the probability of the assembly spot for the first and last contact.

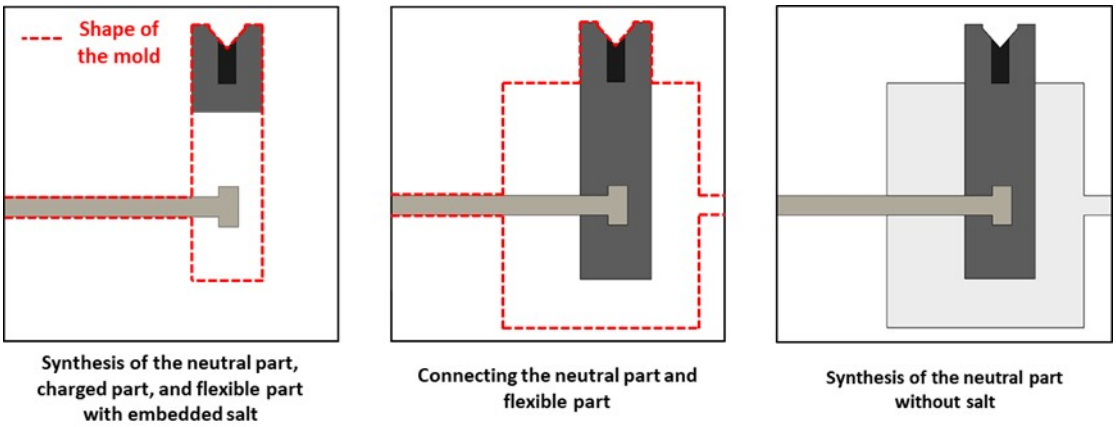

Fig. S12 Synthesis procedure for the hydrogel-based logic gate.

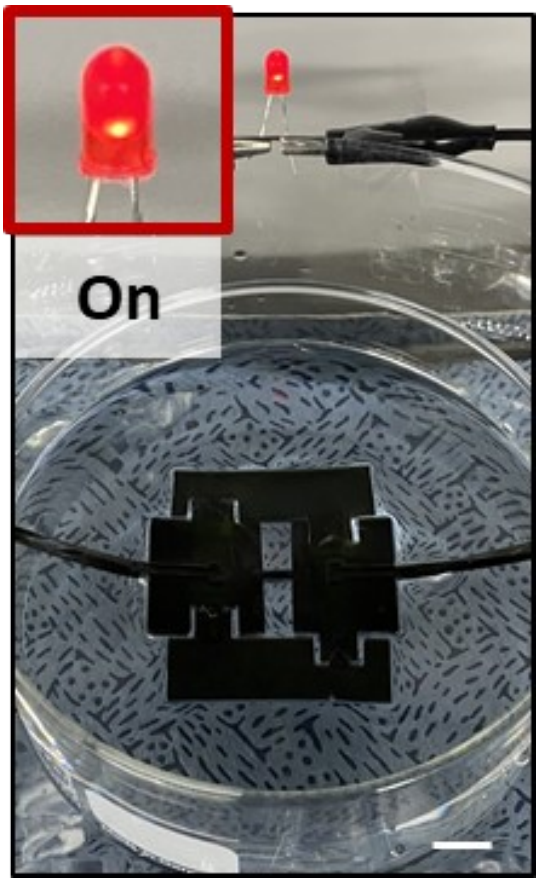

Fig. S13 Experimental image showing the connection of the two complementary building blocks to the main part in the OR logic gate.