Electronic Supplementary Material (ESI) for Biomaterials Science
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Supporting Information for

Programmable polymer-DNA hydrogels with dual input and multiscale response

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Synthetic Methods

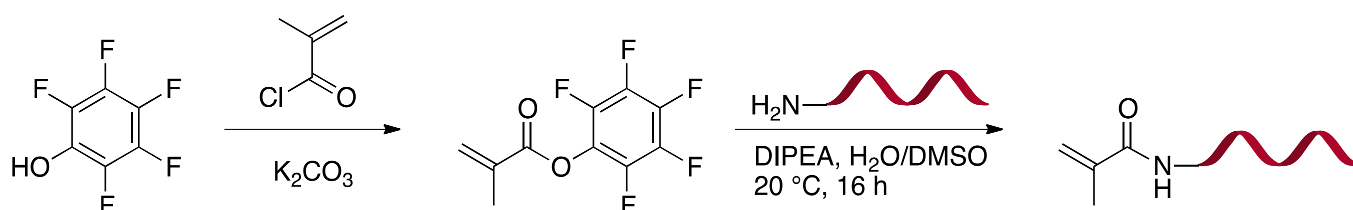

Figure S1: Synthesis of pentafluorophenyl methacrylate and subsequent modification of oligonucleotides.

Characterisation of oligonucleotides and modified oligonucleotides

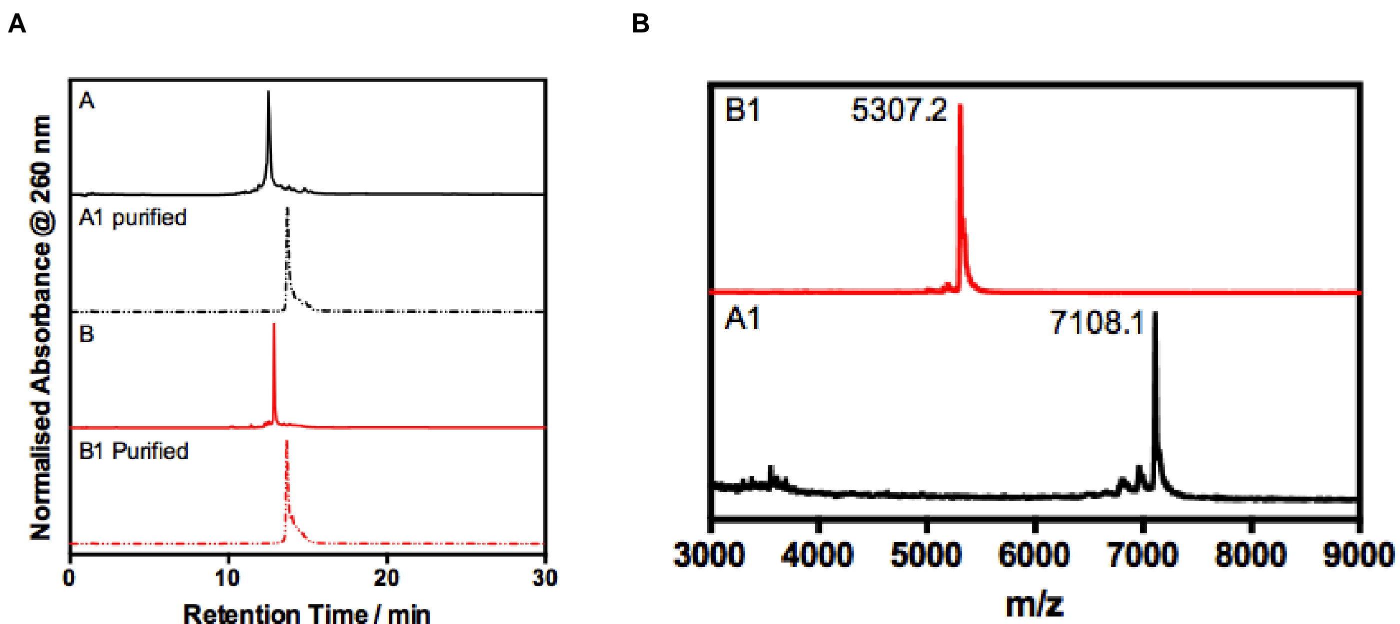

Figure S2. A HPLC chromatograms for the synthesis of methacrylamido oligonucleotides A1 and B1; **B** MALDI-TOF spectra of methacrylamido oligonucleotides A1 and B1 after reverse phase HPLC purification.

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Cryo-Scanning Electron Microscopy

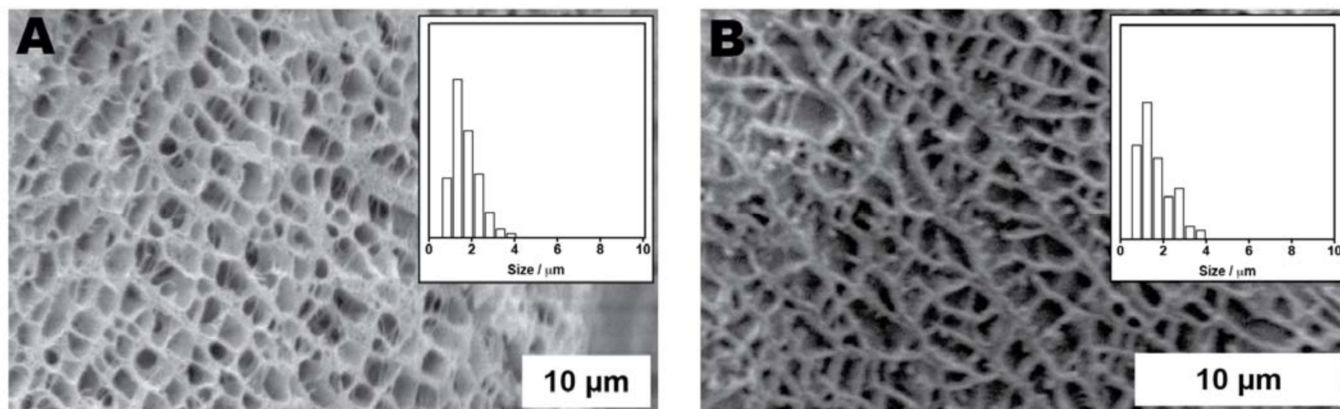

Figure S3: Cryo-scanning electron micrographs and pore size frequency distributions of **A** AAm–BAC–DNA hydrogel after incubation with scrambled oligonucleotide D; **B** AAm–BAC–DNA hydrogel after incubation with water;

Release study of FITC-Dextran loaded AAm–BAC-DNA hydrogels

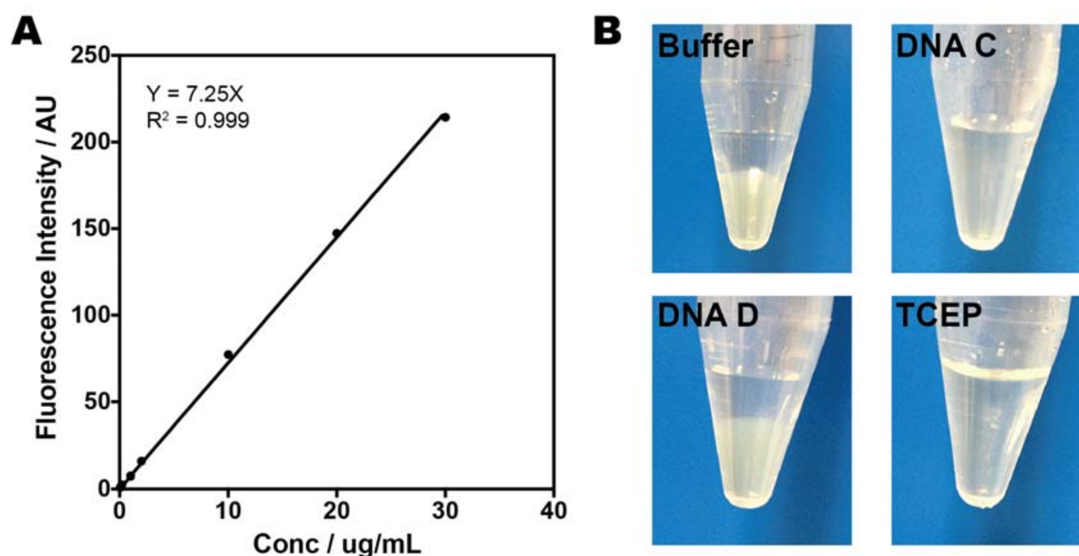

Figure S4. A Standard curve of FITC-Dextran fluorescence versus concentration. **B** Photographs of AAm–BAC–DNA hydrogels after 24h of incubation with DPBS buffer, complementary DNA C, scrambled oligonucleotide D and TCEP•HCl.

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Transport of microparticles through AAm–BAC–DNA hydrogel

AAm–*BAC*–DNA hydrogels loaded with one particle type

To further confirm that the size-gated transport of fluorescent microparticles across the AAm-BAC-DNA hydrogels was induced by directed changes in their crosslinking density, we decided to investigate whether gels loaded with one particle type might exhibit the same transport pattern described for hydrogels treated with a mixture of microparticles. In addition to the experiments discussed in the results section of the main paper, for these assays TCEP and /or the competing strand DNA C were added to the gels *after* the loading of particles to monitor their gating capabilities over time. As expected, the 1 μm Nile Blue marked particles were able to flow across the AAm-BAC-DNA gel due to their reduced dimension (Figure S5). In contrast, the 3 μm FITC and 6μm Rhodamine B labeled particles were detected in the bottom part of the capillary tube only after the addition of DNA C and TCEP respectively (Figure S6 and S7). Although some changes were made to the experimental design across these set of assays, no marked variation in the particle transport was observed.

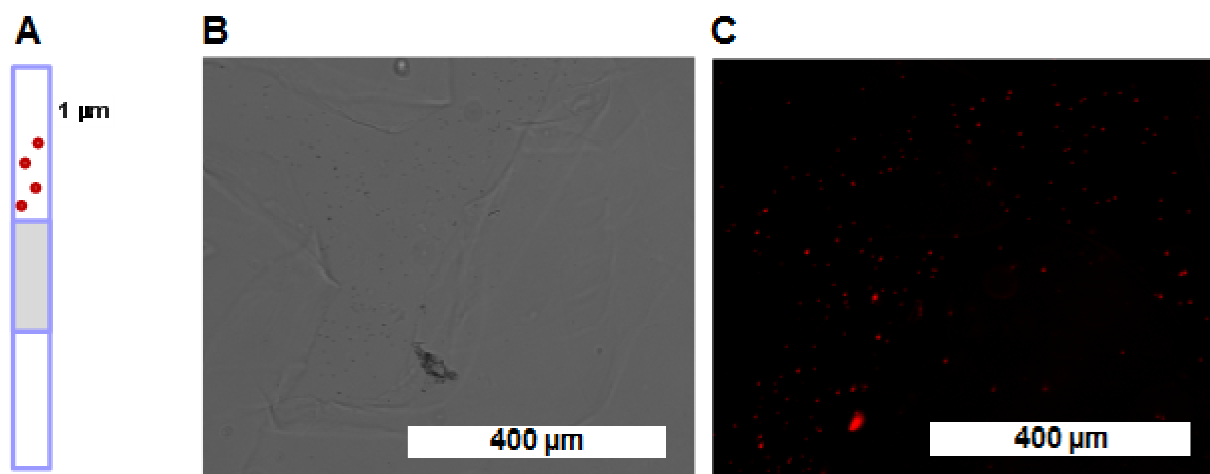

Figure S5: A Schematic of the AAm-BAC-DNA hydrogel loaded with 1 μm Nile-Blue labelled microparticles; **B- C** Optical and fluorescence micrographs of 5 μl aliquot taken from the bottom part of the gel 30 min after the particle loading respectively.

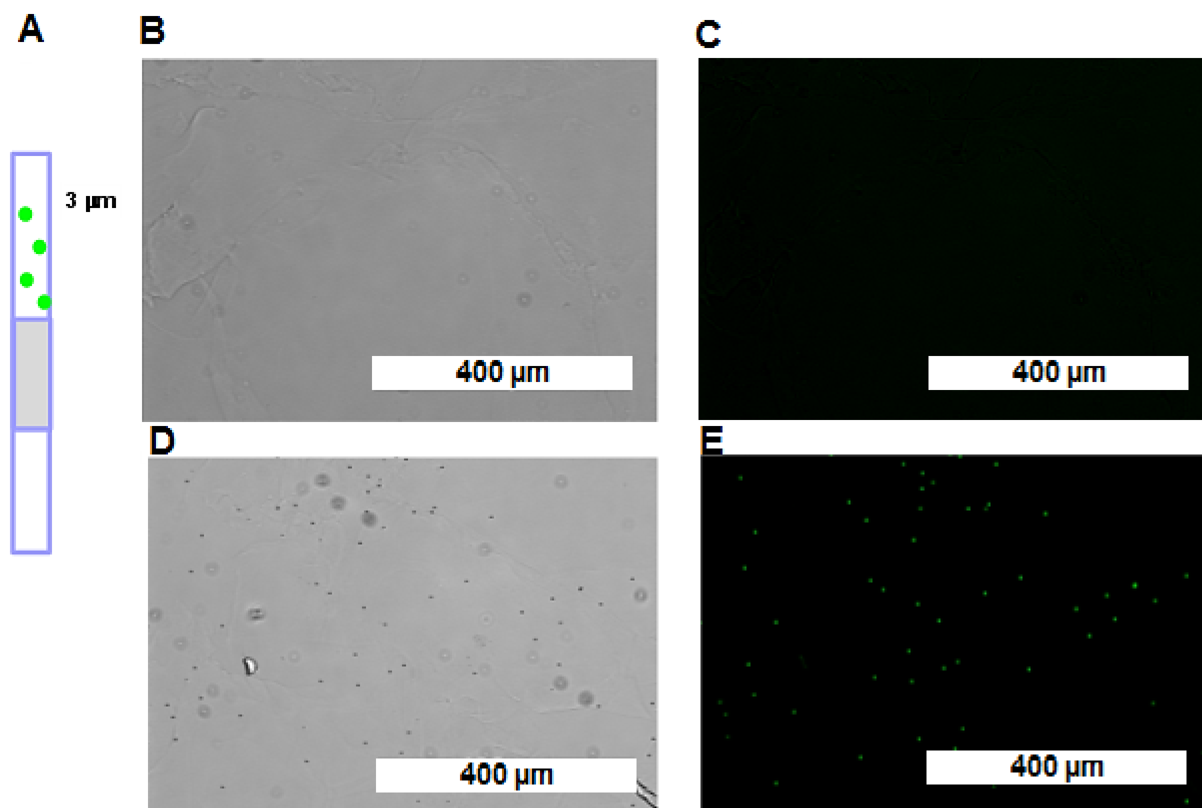

Figure S6: A Schematic of the AAm-BAC-DNA gel loaded with 3 µm FITC marked microparticles; Optical and fluorescence micrographs of 5 µl aliquots taken from the bottom part of the gel 30 min after the particle loading (**B-C**); 1h after the addition of ss-DNA C (**D-E**).

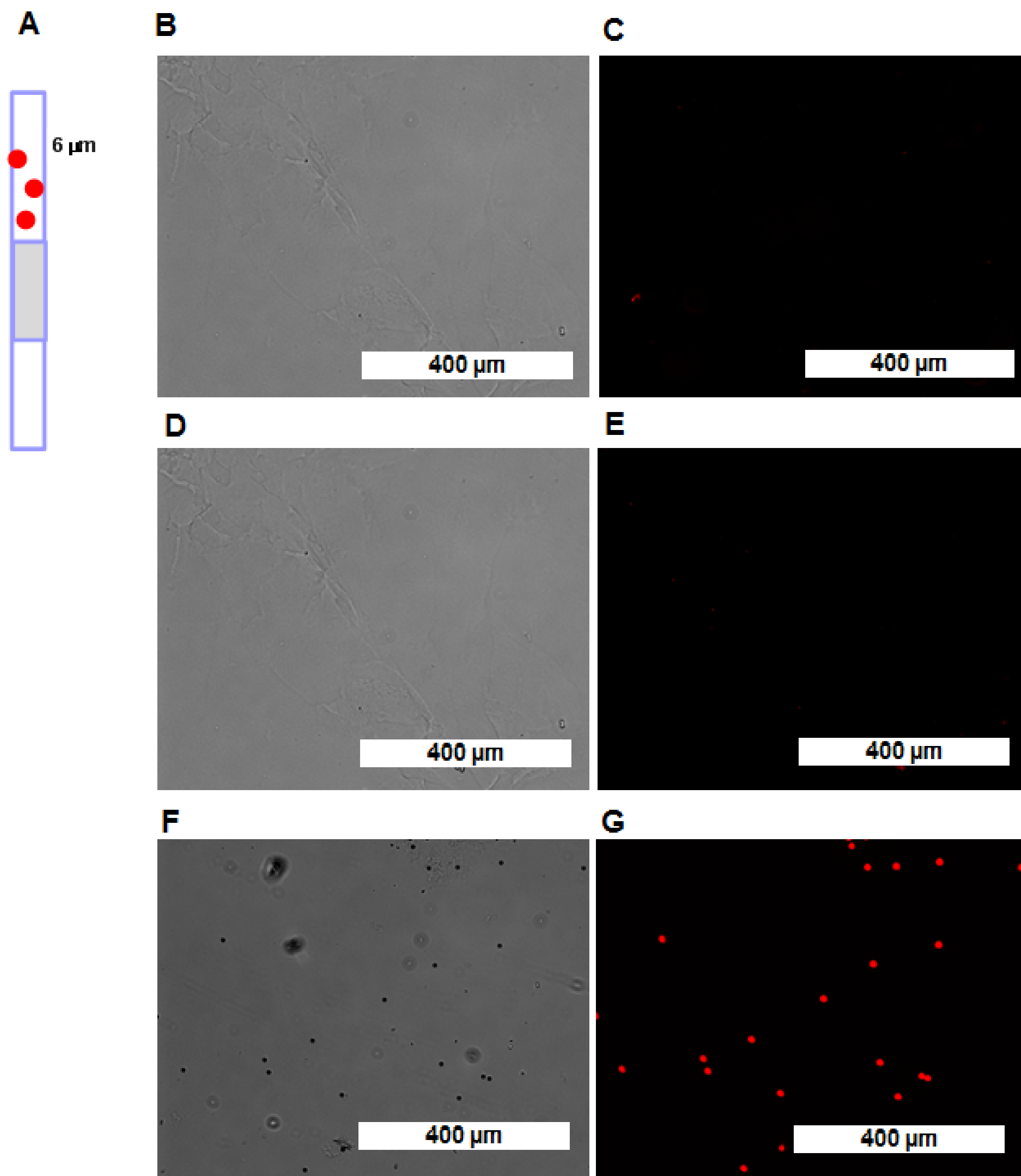

Figure S7: A. Schematic of the AAm-BAC-DNA gel loaded with 6 µm Rhodamine B labeled microparticles; Optical and fluorescence micrographs of 5 µl aliquots taken from the bottom part of the gel 30 min after the particle loading (**B-C)**; 1h after the addition of ss-DNA C (**D-E**); 1h after the addition of TCEP HCl (**F-G**).

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[1] C. A. Schneider, W. S. Rasband, K. W. Eliceiri, *Nature Methods* **2012**, *9*, 671.