## **Supporting Information**

Towards reversible bioresponsive aptamer-based nanocomposites: ATP binding and removal from DNA-grafted silica nanoparticles.

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Figure S1. Characterization of 120- and 290-SiNP by TEM.

Figure S2. Characterization of 120-SiNP-SH by solid state <sup>13</sup>C NMR.

Figure S3. Characterization of ssDNA1, ssDNA2, and dsDNA1-2.

Figure S4. DLS control experiment for NP interactions.

Figure S5. Cryo-TEM of networks after addition of ATP and CTP.

Figure S6. Dilution of the system during the dialysis process in absence of ATP.

Figure S7. TEM of assembly and disassembly of networks over 5 rounds of ATP detection.



Figure S1. TEM micrographs of (a) 120-SiNP and (b) 290-SiNP.



**Figure S2.** <sup>13</sup>C solid state NMR of 120-SiNP-SH. The two peaks appearing at 17.5 and 58.9 ppm are assigned to unhydrolyzed ethoxy groups in the hybrid  $SiO_2$  matrix. The peak at 27.7 ppm is assigned to methylene C atoms 2 and 3, and the peak at 11.6 ppm is due to methylene C atom 1 of attached MPTMS.



**Figure S3.** Characterization of ssDNA1, ssDNA2, and the resulting dsDNA1-2. (a) Fluorescence spectroscopy: (**■**) ssDNA1, (**□**) ssDNA2, and (**●**) dsDNA1-2. Fluorescence of EB alone is given as reference (dashed line). (b) Gel electrophoresis.  $[dsDNA] = 100 \text{ nM}, [EB] = 1\mu \text{M}.$ 



**Figure S4.** DLS control experiment for NP interactions: 5 nM DNA (*for 290-SiNP-DNA2*), 25 nM DNA (*for 120-SiNP-DNA2*) in Tris buffer, pH 8.3.



**Figure S5.** Cryo-TEM of SiNP-DNA networks (a) in absence of target, and after addition of (b) ATP, and (c) CTP. [DNA] = 25 nM, [ATP] = [CTP] = 0.5 mM.



**Figure S6.** Characterization of the dilution of the system during the dialysis process in absence of ATP: (a) in absence and (b) presence of SiNPs. Fluorescence intensity at 650 nm before dialysis (**•**), and after 1 day ( $\Box$ ), 2 days (**•**), 3 days (**•**), and 4 days dialysis (**•**). [dsDNA] = 100 nM, [EB] = 1µM. Fluorescence of EB alone, is given as reference (dashed-dotted line).



**Figure S7** TEM of (a1, b1,c1, d1, e1) re-assembly of networks after dialysis, and (a2, b2,c2, d2, e2) their disassembly upon ATP addition (5 mM), over 5 rounds of ATP detection.