# Supporting information

## Experiment section

### Materials and methods

Tetraethyl orthosilicate (TEOS), and 3-aminopropyltriethoxysilane (APTES) were purchased from Sigma-Aldrich. N-cetyltrimethylammonium bromide (CTAB), 1-ethyl-3-(3-dimethylaminopropyl) carbodiimide (EDC), N-Hydroxysuccinimide (NHS) and succinic anhydride were obtained from Alfa Aesar. All other chemicals were used as received without further purification. Water used in all experiments was purified by a Millipore filtration system. DNA sequences used:

A: 5'-GAAGA CTCGT AATGT GAAAC CG-3',

B: 5'-CACAT TACGA GTCTT CGTGG CATAT CACTCTTGGA G-3',

C: 5'-GGGGT CCGCT ATAAA CACCT CCAAG AGTGA TATGCCAC-3',

D: 5'-GTGTT TATAG CGGAC CCC-$NH_2$-3',

D2: 5'-GTGTT TATAG CGGAC CCC- 3',

A': 5'- CGGTT TCACA TTACG AGTCT TC-3',

B': 5'-CTCCA AGAGT GATAT GCCAC GAAGA C-3',

C': 5'-TTGGA GGTGT TTATA GCGGA CCCC-3'.

Scanning electron microscopy (SEM) was conducted on a Hitachi S-4800 Field Emission Scanning Microscope (acceleration voltage, 10 kV). Transmission electron microscopy (TEM) was conducted on a JEOL JEM-1011 (acceleration voltage, 100 kV). The powder X-ray diffraction (XRD) patterns of the samples were measured on a Bruker D8 FOCUS Powder X-ray Diffractometer using Cu Kα radiation. FT-IR analyses were carried out on a Bruker Vertex 70 FT-IR Spectrometer. N2 adsorption-desorption isotherms were recorded on a Micromeritics ASAP 2020M automated sorption analyzer. Thermogravimetric analyses were carried out on a PerkinElmer Pyris Diamond TG/DTA Analyzer. UV-vis spectroscopy was carried out with a JASCO V-550 UV/vis Spectrometer.

### Synthesis and chemical modification of the MSNs

The carboxylated nanoparticles (MSNs-COOH) were first synthesized according to the literature. Then MSNs-COOH (5 mg) were suspended in $H_2O$ (3 mL) containing EDC (5 mg) and NHS (2 mg), and stirred at room temperature for 8 h. The amine-modified oligonucleotide (strand D) with a solution concentration of 5.0 μM was then added to react overnight to get MSNs-DNA. The unreacted DNA was removed by centrifugation and washing with phosphate-buffered saline (PBS, 20 mM, 50 mM NaCl, pH 7.4).

The purified MSNs-DNA were incubated in the PBS buffer of RhB (0.6 mM, 2.5 mL) for 6 h followed by centrifuging and repeated washing with PBS to remove physisorbed RhB from the exterior surface of the material. Then DNA strand C, B, and A were added in sequence and incubated for 12 h in PBS. The sample was then centrifuged to remove the unattached free DNA.

**Controlled dye release experiments**

For the release experiment, a container whose bottom was covered with a dialysis membrane with a molecular cut-off of 30000 g/mol was used. An image of the container can be seen in Figure S4. The suspension (0.5 mL) of MSNs-DNA-RhB was pipeted into the container, which was subsequently closed by a lid. The closed container is then put onto a cuvette, which is completely filled with PBS buffer. The released dye and wasted DNA strands are able to pass through the applied membrane while the relatively larger particles are retained. The controlled release experiments were performed by addition of different combinations of DNA. UV/vis spectroscopy was used to monitor the amount of released RhB molecules in aqueous solution.

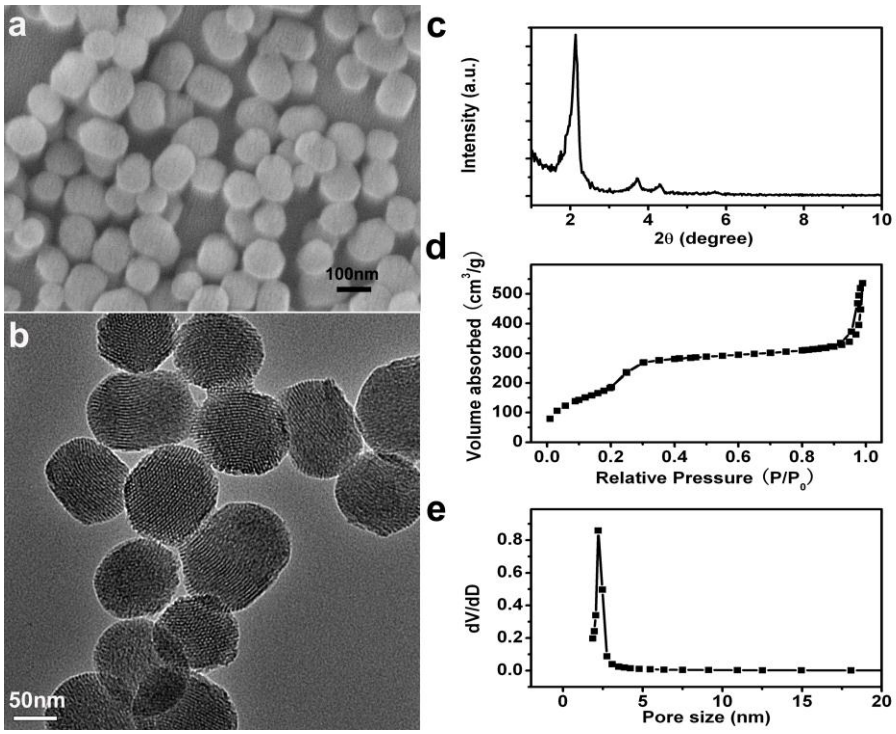


Fig. S1 Characterization of MSNs-COOH. (a) SEM image, (b) TEM image, (c) X-Ray Powder diffraction pattern, (d) nitrogen sorption isotherms, and (e) the corresponding pore size distribution.

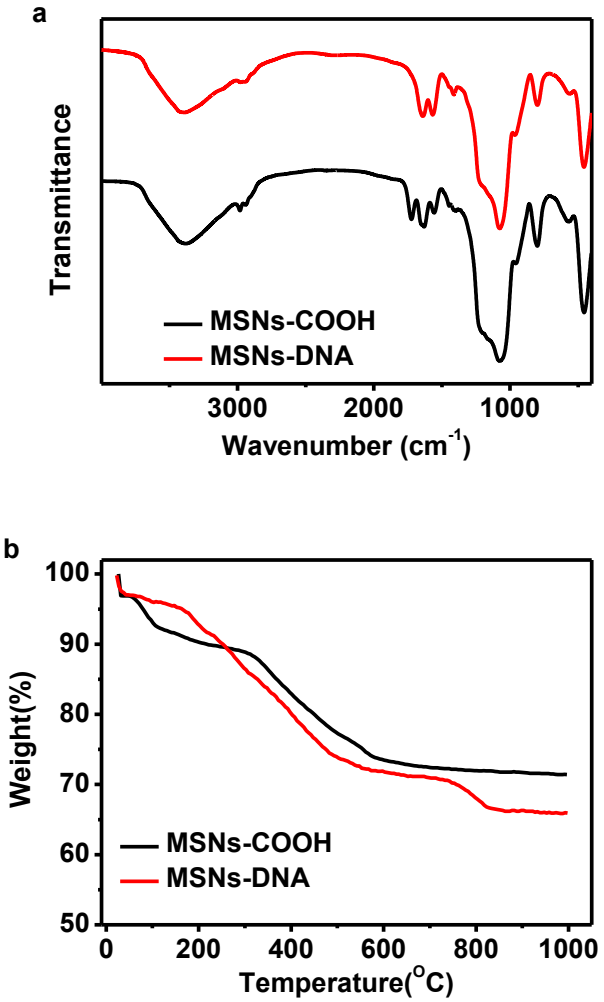


Fig. S2 (a) FTIR spectra of MSNs-COOH and MSNs-DNA. (b) Thermogravimetric analysis of the samples.

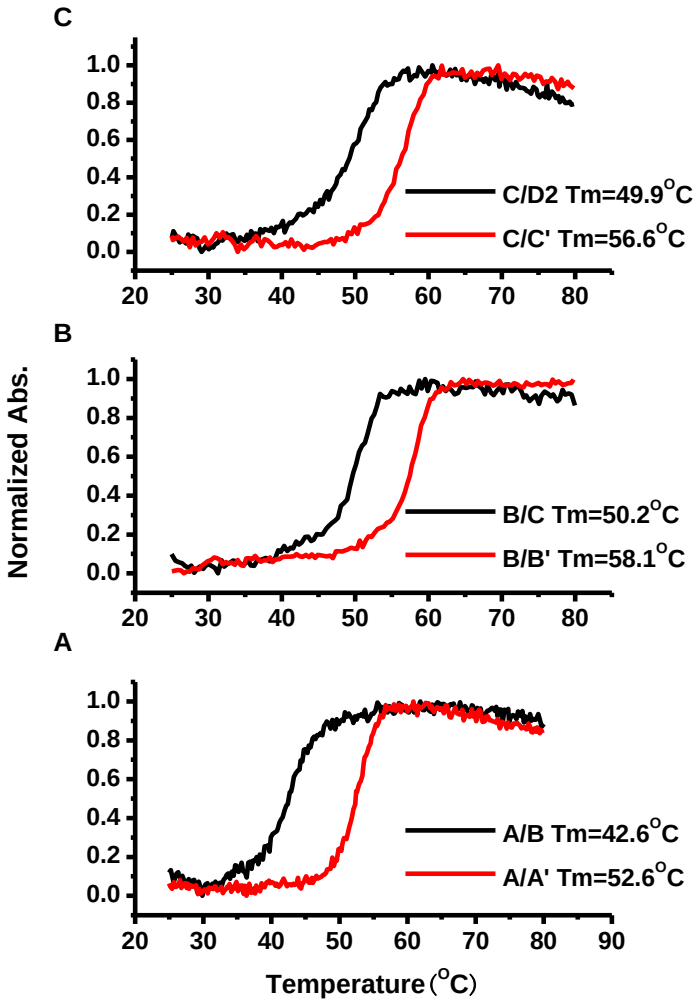


Fig. S3 Thermal melting profiles of samples.

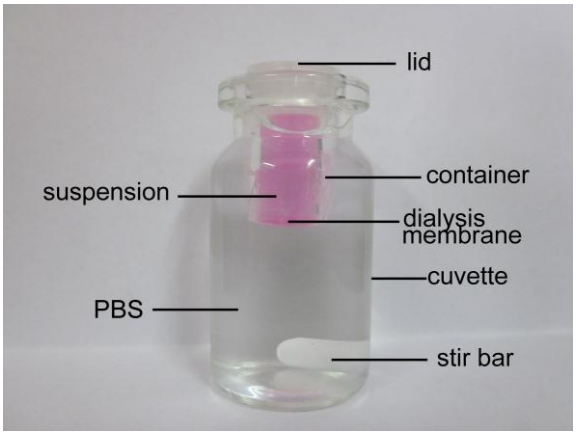


Fig. S4 Release experiment setup featuring a container whose bottom is covered with a dialysis membrane.

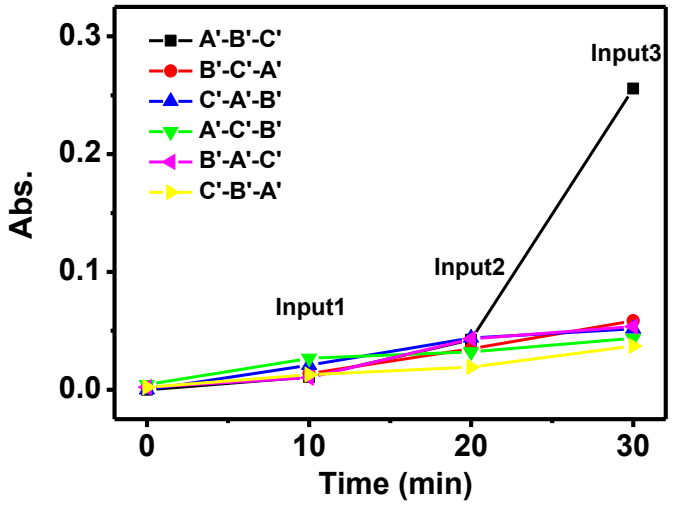


Fig. S5 Absorbance intensities of RhB at 553 nm in the presence of the three input signals with different adding sequences.

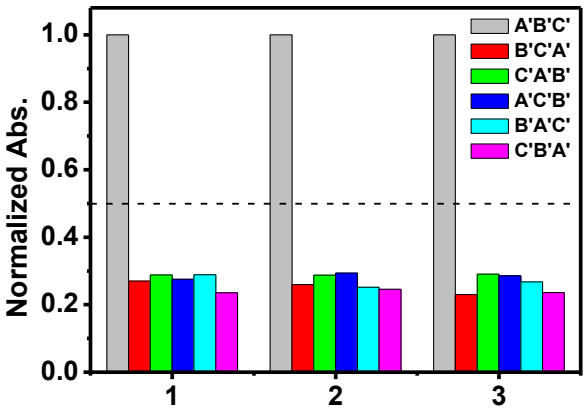


Fig. S6 Normalized absorbance intensities of RhB at 553 nm in the presence of the three input signals with different adding sequences after resetting the system.

In the experiment, the system was separated from the current solution by centrifugation and then was incubated with DNA strand C, B, and A in PBS. Dye release experiment was carried out in fresh PBS. The result demonstrated the system still could implement keypad lock function.