## **Supporting Information**

## **pH Responsive Dual Cargo Delivery from Mesoporous Silica Nanoparticles with a Metal-latched Nanogate**

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Scheme S1. Synthesis and attachment of nanogate machine.



Figure S1. TEM image of a typical batch of synthesized and acid extracted MCM-41 nanoparticles.



**Figure S2.** Powder XRD diffraction of synthesized MCM-41 nanoparticles.



**Figure S3.** IR of surfactant extracted synthesized MSN.



**Figure S4a.** SS <sup>13</sup>C NMR spectrum of iminodiacetic acid modified MSN. Peaks corresponding to IDA are observed in the region >50 ppm. Bare nanoparticles with –OMe terminated silanes do not show peaks in this region.



Figure S4b. SS<sup>29</sup>Si NMR spectrum of iminodiacetic acid modified MSN.



**Figure S5.** Time resolved fluorescent spectrum release profile of Hoechst 33342 cargo from a nickel latched nanogate machine. Solid 2,2'-bipyridine was added as a competitively binding ligand to remove nickel from the iminodiacetic acid nanogates and initiate cargo release. The slow rate of release was due to the low solubility of 2,2'-bipyridine in water.



**Figure S6.** UV-Vis spectra of solution Co/EDTA complexes after simulated loading conditions. The oxidation state of cobalt was determined through spectroscopic methods after mimicking the loading and capping conditions of the nanoparticles. In green: UV-Vis spectra of a Co(II) solution in water. In pink: UV-Vis spectra of 5 mM Co(II)EDTA. In blue: Co(III)EDTA complex (Xue, Y.; Traina, S. J. *Environ. Sci. Technol.* **1996**, 30, 1975-1981). In black: UV-VIS spectra of 10 mM cobalt/EDTA complex after undergoing similar conditions as used in the loading/capping procedures. After this process, the cobalt is bound to EDTA as shown in the increase in absorbance at 470 nm, and most of the cobalt/EDTA is still in the +2 oxidation state as indicated by the absence of a peak at 370 nm.



**Figure S7.** Experimental setup for time-resolved fluorescence spectroscopy. An excitation beam is aimed at the solution supernatant, exciting any dye molecules present. A CCD detector cooled to liquid nitrogen temperatures integrates intensities at a specified wavelength in real-time to generate a typical release profile curve.