Supporting Information

Synthesis of MOF. DMF/H₂O (6 mL, 5:1) solution of $Zn(NO_3)_2 \cdot 6H_2O$ (0.1 mmol), L(0.1 mmol), H₃BTC(0.1 mmol) in a ratio of 1:1:1 was heated at 120°C in a Teflon reactor for 2 days, then cooled to the room temperature at 3°C/h. Pure block crystals were filtrated and drying naturally with the yield of 92% based on Zn.

Uranium Uptake Experiments. The uranium uptake was carried out by the batch method. For example, uptake experiments were performed at controlled pH (1-5) and 298K by adding 10mg MOF material to 10 mL of a uranyl solution in a 50 mL high-density Polymer polypropylene tube (previously cleaned with 5% HNO₃ and rinsed three times with ultrapure water). The solution pH was adjusted to the desired value by HCl (1M)/NH₃H₂O(1:1) and determined by pH meter. After shaking for predesigned time with fixed temperature in the ZWY-240 Constant shaking incubator, the suspension was centrifuged at 10,000 rpm for 10 min, then the sorbent was separated by centrifugation and the rest solution is analyzed for U concentration by spectrophotometer, and further confirmed by inductively coupled plasma-mass spectroscopy (ICP-MS). Arsenazo III dye was used as the color-developing agent for U, giving an intense absorption band at 650 nm. Before the determination, the solution was diluted 2.5-100 times to make sure that the finally uranium concentration meets the standard curve in the range of 0.1-3 mg/L. For extra low ion concentration (\leq ppb) like that of artifical seawater is determined by inductively coupled plasma-mass spectroscopy (ICP-MS, NexION 300X).

Characterization Techniques. Thermogravimetric analysis (TGA) was performed by a TGA Q500 thermal analysis system. All TGA experiments were performed under a N₂ atmosphere from 40-800°C at a rate of 5°C /min. Data were analyzed using the TA Universal Analysis software package. X-ray powder diffraction were collected by a Bruker AXS D8 Discover powder diffractometer at 40 kV, 40 mA for Cu K α , ($\lambda = 1.5406$ Å). The simulated powder patterns were calculated by Mercury 1.4. The purity of the bulk products were determined by comparison of the simulated and experimental PXRD patterns. The generator setting is 40 kV and 40 mA. Fourier transformed infrared (FT-IR) spectra of the samples were recorded on a Nicolet-380 Fourier-Transform infrared spectrometer using the KBr pellet method. SEM and EDS measurements and elemental distribution mappings were carried out using a Hitachi S-4800 microscope.