Highly Efficient Uptake of Cs\textsuperscript{+} by Robust Layered Metal Organic Frameworks with a Distinctive Ion Exchange Mechanism

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Section S1. Ion-exchange experiments

In the kinetic experiments, 50 mg of grinded polycrystalline powder samples of 1 or 2 were weighed into 50 mL of 6.5 mg/L and 3.9 mg/L of Cs\(^+\) aqueous solution, respectively. The mixture was kept under magnetic stirring. The suspensions were sampled at different time intervals (0, 1, 2, 5, 30, 120, 180, 420, 600, 720 min for 1; 0, 1, 2, 5, 10, 20, 30, 60, 120, 180, 300, 420 min for 2), which were filtered by the 0.22 μm Millipore filter on the 2 mL syringe, and then diluted with ultrapure water to meet the concentration range of the test instrument. The concentrations of Cs\(^+\) were analyzed by ICP-MS (Tables S7 and S8).

The adsorption isotherm experiments of Cs\(^+\) ion exchange with two compounds were carried out as follows: the grinded polycrystalline powder samples of 1 and 2 were added into solutions with different concentrations of Cs\(^+\) ions, respectively. The \(V/m\) values of all samples were 1000 mL/g (\(V = 10\) mL, \(m = 10\) mg). The adsorption isotherm experiments lasted about 10 h with continuous stirring at room temperature. Then, the filtered suspensions were analyzed by ICP-MS (Tables S9 and S10).

The competitive experiments of 1 and 2 under the individual or coexisting K\(^+\), Na\(^+\), Mg\(^{2+}\), Ca\(^{2+}\) and Sr\(^{2+}\) were also investigated. 1 and 2 were added to the as-prepared aqueous solutions, respectively. The \(V/m\) ratio is also 1000 mL/g (\(V = 10\) mL, \(m = 10\) mg). The mixed solutions were stirred for 10 h at room temperature. The concentrations of metal ions were analyzed by ICP-MS or ICP-OES (Tables S12-S16). The simulated Cs\(^+\) ion contaminated solutions were prepared with tap water (Fuzhou, Fujian), Minjiang River (Fuzhou, Fujian), river water (Longyan, Fujian), simulated groundwater and sea water (Xiamen, Fujian), containing low concentration of Cs\(^+\). Then, ion-exchange experiments were done (Tables S17 and S18).

The ion-exchange experiments of 1 and 2 before and after irradiation were also carried out. The pristine powder samples were irradiated by 100 kGy \(\gamma\), and 200 kGy \(\gamma\) irradiations. 10 mg of pristine 1 and 2 samples and its irradiated samples were added to 10 mL of Cs\(^+\) aqueous solutions, respectively. The concentrations of Cs\(^+\) ions in filtered solutions were determined by ICP-MS. (Table S19).

In elution and reuse experiments, the raw material for the cycle experiments was firstly prepared: 300 mg of 1 was added to 150 mL of 3000 mg/L Cs\(^+\) ion aqueous solution in a glass bottle and the mixed solutions was shaken for 10 h at room temperature. And the solid product was washed for several times with deionized water, anhydrous ethanol and dried naturally. Then the Cs\(^+\)-exchanged product of 1 was mixed with 150 mL of 0.3 M KCl solution and shaken for 10 h. After that, the solid sample was washed and dried naturally, namely 1-K. The reusability of 1-K was evaluated for three cycles by adsorbing Cs\(^+\) solutions with two different concentrations, and then eluting with 0.3 M KCl solution. The concentrations of Cs\(^+\) ions in solutions were measured after each adsorption (Table S20).

The stability experiments of 1 in solutions with various pH values were carried out. The HNO\(_3\) or NaOH aqueous solutions were prepared with different pH (2.98-12.08) values. 15 mg of polycrystalline powder for 1 were soaked into 15 mL above solutions, respectively. The soaking process lasted 10 h at room temperature, and the \(V/m\) values of all the samples were 1000 mL/g (\(V = 15\) mL, \(m = 15\) mg). Then solid samples were analyzed by PXRD. The concentrations of leaching In were determined by ICP-MS on XSerise II (Table S6).
**Section S2. Crystal structures details**

**Table S1. Crystallographic data and refinement details for compounds.**

| Compounds | 1 | 1-Cs | 1-K | 2 | 2-Cs |
|-----------|---|------|-----|---|------|
| Empirical formula | C$_{21}$H$_{12}$InN$_4$O$_{10}$ | C$_{21}$H$_{12}$InN$_4$O$_{13}$ | C$_{21}$H$_{12}$InK$_{2}$N$_{4}$O$_{13}$ | C$_{21}$H$_{12}$InN$_4$O$_{12}$ | C$_{21}$H$_{18}$.84C$_{6}$H$_{7}$:1InO$_{5}$.28 |
| Formula weight | 610.28 | 696.07 | 602.26 | 612.25 | 666.14 |
| Crystal system | Monoclinic | Monoclinic | Monoclinic | Monoclinic | Monoclinic |
| Space group | $P2_1/c$ | $P2_1/c$ | $P2_1/c$ | $C2/c$ | $C2/c$ |
| T/K | 100(2) | 100(2) | 100(2) | 100(2) | 100(2) |
| $\lambda$/Å | 1.54178 | 1.54178 | 1.3405 | 0.71073 | 1.3405 |
| a/Å | 10.0110(15) | 9.8620(3) | 9.8822(3) | 14.6706(14) | 13.0956(4) |
| b/Å | 16.693(3) | 16.6451(4) | 16.8719(6) | 13.2520(13) | 14.7700(4) |
| c/Å | 14.956(3) | 14.3580(5) | 13.9820(5) | 12.5619(13) | 12.4590(4) |
| $\alpha^{\circ}$ | 90 | 90 | 90 | 90 | 90 |
| $\beta^{\circ}$ | $108.289(18)$ | $107.5269(4)$ | $108.884(4)$ | $96.441(9)$ | $100.745(3)$ |
| $\gamma^{\circ}$ | 90 | 90 | 90 | 90 | 90 |
| $V$/Å$^3$ | 2371.2(8) | 2247.52(13) | 2205.76(14) | 2426.8(4) | 2367.59(13) |
| Z | 4 | 4 | 4 | 4 | 4 |
| $D_0$/ Mg·m$^{-3}$ | 1.708 | 2.057 | 1.814 | 1.676 | 1.869 |
| $\mu$/mm$^{-1}$ | 8.535 | 21.490 | 7.256 | 1.041 | 11.494 |
| F(000) | 1240 | 1352 | 1208 | 1240 | 1303 |
| Measured refls. | 23508 | 18646 | 23596 | 6837 | 8625 |
| Independent refls. | 4837 | 4544 | 4912 | 2924 | 2643 |
| $R_{int}$ | 0.0407 | 0.0363 | 0.0355 | 0.0835 | 0.0285 |
| No. of parameters | 353 | 334 | 328 | 217 | 182 |
| $GOF$ | 1.081 | 1.040 | 1.047 | 1.216 | 1.035 |
| $I/\sigma(I)$ | 0.0360, 0.0402, 0.0365, 0.0605 | 0.0916, 0.1007, 0.0943, 0.1363 | 0.0401, 0.0481, 0.0448, 0.0651 | 0.0944, 0.1075, 0.0999, 0.1386 | 0.0944, 0.1075, 0.0999, 0.1386 |
| $wR_2$ (all data) | 0.0944 | 0.1075 | 0.0999 | 0.1386 | 0.0712 |
| CCDC | 2124172 | 2124173 | 2124174 | 2124175 | 2124176 |

$R_1 = \sum \left| F_o \right| - \left| F_c \right| / \sum \left| F_o \right|$. $wR_2 = \left[ \sum \left( w(F_o)^2 + w(F_c)^2 \right)^{1/2} / \sum w(F_o)^2 \right]^{1/2}$
Figure S1. ORTEP plot showing the crystallographically asymmetric units of (a) 1, (b) 1-Cs, (c) 1-K, (d) 2 and (e) 2-Cs. Thermal ellipsoids are given at the 30% probability level. The hydrogen atoms are omitted for clarity.
Figure S2. The coordination modes of $\text{In}^{3+}$ ion (a) and aip$^{2-}$ ligands (b, c) in 1. The coordination mode of In$^{3+}$ ion in 1-Cs (d). The surrounding environment of Cs$^+$ ion (e) and the coordination mode of aip$^{2-}$ ligand (f) in 1-Cs. The surrounding environment of K$^+$ ion (g) and the coordination mode of aip$^{2-}$ ligand (h) in 1-K.

Figure S3. The anionic layer of [In(aip)$_2$]$^{x-}$ (a) and its simplified topological network (b) in 1.

Table S2. Selected bonds length (Å) in 1, 1-Cs and 1-K.

|        | 1          | 1-Cs         | 1-K         |
|--------|------------|--------------|-------------|
|        | In(1)-O(1) | In(1)-O(1)   | Cs(1)-O(1)  | 3.001(4) |
|        | 2.531(3)   | 2.321(4)     |             |           |
|        | In(1)-O(2) | In(1)-O(2)   | Cs(1)-O(2)  | 3.118(4) |
|        | 2.154(3)   | 2.239(3)     |             |           |
|        | In(1)-O(3)#1 | In(1)-O(3)#1 | Cs(1)#4-O(4) | 3.237(4) |
|        | 2.387(3)   | 2.384(4)     |             |           |
|        | In(1)-O(4)#1 | In(1)-O(4)#1 | Cs(1)-O(5)  | 3.173(4) |
|        | 2.205(2)   | 2.199(4)     |             |           |
|        | In(1)-O(5) | In(1)-O(5)   | Cs(1)-O(6)  | 3.237(4) |
|        | 2.122(3)   | 2.142(4)     |             |           |
|        | In(1)-O(7)#2 | In(1)-O(6)   | Cs(1)-O(7)  | 3.184(8) |
|        | 2.333(3)   | 2.552(4)     |             |           |
|        | In(1)-O(8)#2 | In(1)-O(7)#2 | Cs(1)-O(1W) | 3.182(4) |
|        | 2.216(3)   | 2.296(4)     |             |           |
|        |            | In(1)-O(8)#2 | Cs(1)-O(2W) | 3.125(6) |
|        |            | 2.260(3)     |             |           |
|        |            |            | Cs(1)-O(3W) |             |
|        |            |            | Cs(1)-O(3)  |             |

Symmetry transformations used to generate equivalent atoms: 1: #1 x+1, y, z; #2 -x+2, y+1/2, -z+1/2.
1-Cs: #1 x+1, y, z; #2 -x+1, y+1/2, -z+1/2.
1-K: #1 x-1, y, z; #2 -x, y-1/2, -z+1/2; #3 x, -y+1/2, z+1/2;
\textbf{Table S3.} Hydrogen bonding data for 1.

| D-H···A                  | D-H (Å)  | H···A (Å) | D···A (Å) | <(DHA) (°) |
|-------------------------|----------|-----------|-----------|------------|
| N(1)-H(1A)···O(7)#5     | 0.888(10)| 2.195(17) | 3.057(4)  | 163(4)     |
| N(1)-H(1B)···O(1W)#6    | 0.896(10)| 2.58(4)   | 3.141(5)  | 121(4)     |
| N(2)-H(2A)···N(1)#7     | 0.890(10)| 2.428(13) | 3.314(5)  | 173(4)     |
| N(2)-H(2B)···O(1)#8     | 0.890(10)| 2.151(13) | 3.035(5)  | 172(4)     |
| C(14)-H(14)···O(1W)#9   | 0.95     | 2.46      | 3.389(5)  | 164.7      |
| N(3)-H(3A)···O(9)       | 0.893(10)| 1.83(2)   | 2.703(6)  | 165(6)     |
| N(3)-H(3B)···O(1W)      | 0.893(10)| 1.853(18) | 2.731(6)  | 167(6)     |
| C(17)-H(17B)···O(1)     | 0.98     | 2.38      | 3.161(6)  | 135.7      |
| C(17)-H(17C)···O(8)#2   | 0.98     | 2.59      | 3.312(7)  | 130.7      |
| C(21)-H(21B)···O(5)#2   | 0.98     | 2.44      | 3.383(6)  | 162.2      |
| C(21)-H(21C)···O(2)#10  | 0.98     | 2.31      | 3.242(6)  | 158.7      |
| O(1W)-H(1WA)···O(3)     | 0.831(10)| 1.939(12) | 2.766(4)  | 174(6)     |
| O(1W)-H(1WB)···O(6)#11  | 0.829(10)| 2.030(18) | 2.831(5)  | 162(5)     |

Symmetry transformations used to generate equivalent atoms: #1 x+1, y, z; #2 -x+2, y+1/2, -z+1/2; #3 x-1, y, z; #4 -x+2, y-1/2, -z+1/2; #5 -y+3/2, z+1/2; #6 -x+1, -y+2, -z+1; #7 -x+2, y-1/2, -z+3/2; #8 -x+2, -y+1, -z+1; #9 x+1, -y+3/2, z+1/2; #10 -x+2, -y+2, -z+1; #11 x-1, -y+3/2, z-1/2.

\textbf{Figure S4.} The In-L-In angles in (a) 1, (b) 1-Cs, (c) 1-K, (d) 2 and (e) 2-Cs.
Figure S5. (a) A square-like window with one [(CH$_3$)$_2$NH$_2$]$^+$ cation in 2. (b) The anionic layer of [In(hip)$_2$]$^{n-}$ in 2. (c) The packing arrangement of the layers in 2 viewed along the c-axis. DMF, lattice water molecules, and hydrogen atoms are omitted for clarity. (d) The packing arrangement of the layers in 2 viewed along the c-axis, where the [In(hip)$_2$]$^{n-}$ layers are simplified to topological structures. Lattice water molecules are omitted for clarity. (e) A square-like window with one Cs$^+$ ion in 2-Cs. (f) The packing arrangement of the layers in 2-Cs viewed along the c-axis. Cs1B, lattice water molecules, and hydrogen atoms are omitted for clarity. (g) The packing arrangement of the layers in 2-Cs viewed along the c-axis, where the [In(hip)$_2$]$^{n-}$ layers are simplified to topological structures. Cs1B and lattice water molecules are omitted for clarity.

Table S4. Selected bonds length (Å) in 2 and 2-Cs.

|        | 2          | 2-Cs       |
|--------|------------|------------|
| In(1)-O(1) | 2.195(4)   | 2.3893(17) | Cs(1B)-O(1) | 3.50(3)     |
| In(1)-O(1)#1 | 2.195(4)    | 2.3893(17) | Cs(1B)-O(1)#4 | 2.81(4)    |
In(1)-O(2) 2.356(4)  In(1)-O(2) 2.1861(16)  Cs(1)-O(4)#5 3.359(3)
In(1)-O(2)#1 2.356(4)  In(1)-O(2)#1 2.1861(16)  Cs(1)-O(4)#6 3.620(2)
In(1)-O(3)#2 2.395(4)  In(1)-O(3)#2 2.3461(16)  Cs(1B)-O(4)#5 3.08(4)
In(1)-O(3)#3 2.395(4)  In(1)-O(3)#3 2.3461(16)  Cs(1)-O(1W) 3.202(3)
In(1)-O(4)#2 2.208(4)  In(1)-O(4)#2 2.2094(18)  Cs(1)-O(1W)#4 3.414(3)
In(1)-O(4)#3 2.208(4)  In(1)-O(4)#3 2.2094(18)  Cs(1B)-O(3W)#8 3.591(9)
Cs(1)-O(1) 3.082(2)  Cs(1)-O(1W)#4 3.414(3)  Cs(1)-O(3W)#8 3.134(16)
Cs(1)-O(1B) 3.082(2)  Cs(1)-O(1W)#4 3.414(3)  Cs(1)-O(4W)#8 3.134(16)

Symmetry transformations used to generate equivalent atoms:

2: #1 x+1, y, z+3/2; #2 x-1/2, y-1/2, z; #3 x+3/2, y-1/2, z+3/2.

2-Cs: #1 x+1, y, z+1/2; #2 x-1/2, y-1/2, z+1/2; #3 x+1/2, y+1/2, z; #4 x+1/2, y+1/2, z; #5 x+1/2, y+1/2, z+1/2; #6 x, y+1, z+1/2; #8 x, y, z.

Table S5. Hydrogen bonding data for 2.

| D-H···A       | D-H (Å) | H···A (Å) | D···A (Å) | <(DHA) (°) |
|--------------|---------|-----------|-----------|------------|
| O(5)-H(5A)···O(3)#5 | 0.818(10) | 1.916(13) | 2.733(6)  | 176(7)     |
| C(5)-H(5)···O(3)#5  | 0.95    | 2.62      | 3.298(7)  | 128.9      |
| C(9)-H(9A)···O(2)#1 | 0.95    | 2.61      | 3.489(19) | 153.7      |
| C(11)-H(11B)···O(1)#6 | 0.98    | 2.55      | 3.46(4)   | 154.9      |
| N(2)-H(2A)···O(1)  | 0.91    | 2.32      | 2.958(14) | 126.7      |
| N(2)-H(2A)···O(4)#2 | 0.91    | 2.21      | 3.067(16) | 156.9      |
| C(12)-H(12C)···O(2)#7 | 0.98    | 2.53      | 3.15(2)   | 121.2      |
| C(13)-H(13C)···O(3)#7 | 0.98    | 2.49      | 3.421(19) | 158.8      |
| O(7)-H(7B)···O(4)  | 0.821(10)| 2.58(13)  | 3.174(15) | 130(15)    |

Symmetry transformations used to generate equivalent atoms:

#1 x+1, y, z+3/2; #2 x-1/2, y-1/2, z; #3 x+3/2, y-1/2, z+3/2; #5 x, y+1, z+1/2; #6 x-1/2, y+1/2, z; #7 x+1/2, y+1/2, z+1/2.

Section S3. Characterization of products before and after Cs⁺ ion exchange
Figure S6. The simulated and as-synthesized PXRD patterns of 1 (a); 1-Cs (b); 1-K (c); 2 (d); 2-Cs (e).

Figure S7. (a) PXRD patterns of 1 simulated, as-synthesized, and after soaked in solutions with various pH values for 10 h. (b) The leaching percentages of In for 1 soaked in different pH solutions for 10 h.

Table S6. Dissolution concentrations and leaching percentages of In for 1 after soaked in different pH solutions ($V/m = 1000$ mL/g, contact time = 10 h).

| pH   | Dissolution concentration of In (mg/L) | Leaching percentage of In (%) |
|------|---------------------------------------|-------------------------------|
| 2.98 | 0.0322                                 | 0.01711                       |
| 4.15 | 0.0254                                 | 0.0135                        |
| 5.28 | 0.0234                                 | 0.01244                       |
| 6.66 | 0.0962                                 | 0.05113                       |
| 7.65 | 0.1088                                 | 0.05783                       |
| 8.91 | 0.0936                                 | 0.04975                       |
| 10.13| 0.302                                 | 0.16052                       |
| 11.05| 0.0594                                 | 0.03157                       |
| 12.06| 0.046                                 | 0.02445                       |

Figure S8. Thermogravimetric curves (a) for 1, 1-Cs and 1-K; (b) for 2 and 2-Cs.
Figure S9. Optical absorption spectra (a) for 1, 1-Cs, and 1-K; (b) for 2 and 2-Cs.

Figure S10. EDS diagrams of 1-Cs (a) and 2-Cs (b).

Figure S11. X-ray photoelectron spectra of nitrogen (a) for 1 and 1-Cs; (b) for 2 and 2-Cs.
Section S4. Kinetic and capacities studies for Cs⁺ ion exchange

Equations for ion exchange analysis

The removal rate $R$ (%) is to evaluate the removal percentage of ion (equation S1). The pseudo-first-order and pseudo-second-order kinetics could be expressed by equations S2 and S3:

$$ R = \frac{(C_0 - C_e)}{C_e} \times 100\% \quad (S1) $$

$$ \log (q_e - q_t) = \log q_e - \frac{k_1}{2.303} t \quad (S2) $$

$$ \frac{t}{q_t} = \frac{1}{k_2 q_e^2} + \frac{t}{q_e} \quad (S3) $$

Where $C_0$ (mg/L) and $C_e$ (mg/L) represent the initial and equilibrium concentrations of cesium at adsorption experiment. $q_e$ and $q_t$ are the adsorption capacities for cesium (mg/g) at equilibrium and time $t$, respectively. The pseudo-first-order and pseudo-second-order rate constants are represented by $k_1$ (min⁻¹) and $k_2$ (g/mg min), respectively. The slope and intercept of the linear graph of $\log(q_e - q_t)$ vs $t$ can be used to calculate the values of $q_e$ and $k_1$, respectively. The slope and intercept of the linear graph of $t/q_t$ vs $t$ can be used to calculate the values of $q_e$ and $k_2$, respectively.

The amount of cesium $q$ (mg/g) absorbed by absorbents can be calculated by equation S4. Evaluation of saturation capacity $q_m$ (mg/g) can be calculated by the Langmuir adsorption isotherms and Langmuir-Freundlich adsorption isotherms, as depicted in equations S5 and S6.

$$ q = \frac{(C_0 - C_e)V}{m} \quad (S4) $$

$$ q = q_m \frac{bC_e}{1 + bC_e} \quad (S5) $$

$$ q = q_m \frac{\left(\frac{bC_e}{1 + (bC_e)^n}\right)^{\frac{1}{n}}}{1 + (bC_e)^n} \quad (S6) $$

Where $C_0$ (mg/L) and $C_e$ (mg/L) represent the initial and equilibrium concentrations of cesium at adsorption experiment, $V$ (mL) and $m$ (g) are the volume of the solution and the mass of the adsorbent in the ion exchange experiment, respectively. $q_m$ is the maximum adsorption capacity (mg/g). $b$ (L/mg), related to the free energy of exchange, is a Langmuir constant. $C_e$ (mg/L) is the equilibrium concentration, $n$ is a Freundlich constant.

The affinity and selectivity of adsorbents to cesium ions can be measured by distribution coefficient ($K_d$). It could be described by equation S7.

$$ K_d = \frac{V}{m} \frac{(C_0 - C_e)}{C_e} \quad (S7) $$

The separation factor $SF$ is a scale for determining whether the medium can separate two ions from each other, as depicted in equation S8:

$$ SF = \frac{K_d^A}{K_d^B} \quad (S8) $$

Where, $K_d^A$ and $K_d^B$ represent the distribution coefficients of A and B ions, respectively.
Table S7. The data for the concentrations of Cs⁺ (C) and the relative amounts of Cs⁺ removed (R) for 1 at the different time in kinetics experiments.

| Time (minutes) | C₅Cs (mg/L) | R₅Cs (%) |
|---------------|-------------|----------|
| 0             | 6.50        | 0        |
| 1             | 0.46        | 92.92    |
| 2             | 0.29        | 95.54    |
| 5             | 0.25        | 96.15    |
| 30            | 0.24        | 96.31    |
| 120           | 0.32        | 95.08    |
| 180           | 0.33        | 94.92    |
| 420           | 0.39        | 94.00    |
| 600           | 0.42        | 93.54    |
| 720           | 0.44        | 93.23    |

Table S8. The data for the concentrations of Cs⁺ (C) and the relative amounts of Cs⁺ removed (R) for 2 at the different time in kinetics experiments.

| Time (minutes) | C₅Cs (mg/L) | R₅Cs (%) |
|---------------|-------------|----------|
| 0             | 3.90        | 0        |
| 1             | 0.42        | 89.23    |
| 2             | 0.42        | 89.23    |
| 5             | 0.36        | 90.77    |
| 10            | 0.33        | 91.54    |
| 20            | 0.32        | 91.79    |
| 30            | 0.34        | 91.28    |
| 60            | 0.38        | 90.26    |
| 120           | 0.51        | 86.92    |
| 180           | 0.52        | 86.67    |
| 300           | 0.58        | 85.13    |
| 420           | 0.60        | 84.62    |
Figure S12. The fitting results with pseudo-first-order kinetic model (a, c) and pseudo-second-order kinetic model (b, d) for the Cs⁺ ions removal kinetics of 1 and 2, respectively.

Table S9. Adsorption capacities for 1 at the different initial Cs⁺ concentrations.

| $C_0^{Cs}$ (mg/L) | $C_e^{Cs}$ (mg/L) | $q^{Cs}$ (mg/g) |
|-------------------|-------------------|-----------------|
| 24                | 4                 | 20              |
| 80                | 8.6               | 71.4            |
| 140               | 38                | 102             |
| 274.5             | 63                | 211.5           |
| 460               | 210               | 250             |
| 506               | 255.5             | 250.5           |
| 630               | 390               | 240             |
| 830               | 590               | 240             |

Table S10. Adsorption capacities for 2 at the different initial Cs⁺ concentrations.

| $C_0^{Cs}$ (mg/L) | $C_e^{Cs}$ (mg/L) | $q^{Cs}$ (mg/g) |
|-------------------|-------------------|-----------------|
| 24                | 7.0               | 17              |
| 44                | 8.7               | 35.3            |
| 80                | 24                | 56              |
| 140               | 46                | 94              |
| 200               | 78                | 122             |
| 460               | 250               | 210             |
| 1105              | 819.5             | 285.5           |
| 2620              | 2350              | 270             |
Table S11. Isotherm fitting parameters for the capture of Cs⁺ ions by 1 and 2 (V/m = 1000 mL/g, room temperature, 10 h contact time).

| Compounds | Langmuir model | Langmuir-Freundlich model |
|-----------|----------------|--------------------------|
|           | q_m (mg·g⁻¹)  | b (L·mg⁻¹) | R² | q_m (mg·g⁻¹) | b (L·mg⁻¹) | n | R² |
| 1         | 270.86         | 0.03028 | 0.9229 | 262.89 | 0.03186 | 0.8752 | 0.91037 |
| 2         | 297.67         | 0.00987 | 0.98714 | 295.82 | 0.01006 | 0.97771 | 0.98466 |

Section S5. Studies on competitive ion exchange

Table S12. The results for the Cs⁺-exchange of 1 under individual competitive excessive alkali, and alkaline-earth ions (V/m = 1000 mL/g, room temperature, 10 h contact time).

| Compounds | Initial M/Cs molar ratio | C_0^M (mg/L) | C^M (mg/L) | C_0^Cs (mg/L) | C^Cs (mg/L) | R^Cs (%) | Kd^Cs (mL/g) | R^M (%) | Kd^M (mL/g) |
|-----------|--------------------------|--------------|------------|---------------|------------|---------|-------------|---------|-------------|
| K/Cs      | 19.95                    | 12.53        | 8.231       | 2.135         | 0.217      | 89.84   | 8.84 × 10³  | 34.31   | 522.29      |
| Na/Cs     | 92.96                    | 22.52        | 22.38       | 1.4           | 0.22       | 84.29   | 5.36 × 10³  | 0.62    | 6.26        |
| Ca/Cs     | 27.67                    | 18.525       | 18.47       | 2.22          | 0.2325     | 89.53   | 8.55 × 10³  | 0.3     | 2.98        |
| Mg/Cs     | 62.22                    | 16.16        | 15.8        | 1.42          | 0.24       | 83.10   | 4.92 × 10³  | 2.23    | 22.781      |

Table S13. The results for the Cs⁺-exchange of 2 under individual competitive excess alkali and alkaline-earth ions (V/m = 1000 mL/g, room temperature, 10 h contact time).

| Compounds | Initial M/Cs molar ratio | C_0^M (mg/L) | C^M (mg/L) | C_0^Cs (mg/L) | C^Cs (mg/L) | R^Cs (%) | Kd^Cs (mL/g) | R^M (%) | Kd^M (mL/g) |
|-----------|--------------------------|--------------|------------|---------------|------------|---------|-------------|---------|-------------|
| K/Cs      | 46.69                    | 19.78        | 16.8       | 1.44          | 0.24       | 83.33   | 5.0 × 10³   | 15.06   | 177.38      |
| Na/Cs     | 72.71                    | 21.424       | 21.252     | 1.7035        | 0.4191     | 75.40   | 3.06 × 10³  | 0.80    | 8.09        |
| Ca/Cs     | 45.46                    | 19.74        | 18.54      | 1.44          | 0.28       | 80.56   | 4.14 × 10³  | 6.08    | 64.72       |
| Mg/Cs     | 36.75                    | 18.18        | 17.11      | 2.705         | 0.475      | 82.44   | 4.69 × 10³  | 5.83    | 62.54       |

Table S14. The results for the Cs⁺-exchange of 1 with competitive Sr²⁺ ions (V/m = 1000 mL/g, room temperature, 10 h contact time).

| Initial Sr/Cs molar ratio | C_0^Sr (mg/L) | C^Sr (mg/L) | C_0^Cs (mg/L) | C^Cs (mg/L) | R^Cs (%) | R^Sr (%) | Kd^Cs (mL/g) | Kd^Sr (mL/g) | SF_CsSr |
|---------------------------|---------------|------------|---------------|------------|---------|---------|-------------|-------------|---------|
| 0.03                      | 4.839         | 4.821      | 237           | 118        | 50.21   | 0.37    | 1.01 × 10³  | 3.73        | 270.78  |
| 0.11                      | 6.34          | 5.48       | 87            | 3.8        | 95.63   | 13.56   | 2.19 × 10⁴  | 156.93      | 139.52  |
| 1.69                      | 5.6           | 5.32       | 5.02          | 0.66       | 86.85   | 5.00    | 6.61 × 10³  | 52.63       | 125.59  |
| 15.29                     | 51.4          | 49.3       | 5.1           | 0.64       | 87.45   | 4.09    | 6.97 × 10³  | 42.60       | 163.6   |
| 75.55                     | 254           | 250        | 5.1           | 0.84       | 83.53   | 1.57    | 5.07 × 10³  | 16         | 316.96  |
Table S15. The results for the Cs\(^+\)-exchange of 2 with competitive Sr\(^{2+}\) ions (V/m = 1000 mL/g, room temperature, 10 h contact time).

| Initial Sr/Cs molar ratio | C\(_0\)Sr (mg/L) | C\(_e\)Sr (mg/L) | C\(_0\)Cs (mg/L) | C\(_e\)Cs (mg/L) | R\(_{Cs}\) (%) | R\(_{Sr}\) (%) | K\(_d\)Cs (mL/g) | K\(_d\)Sr (mL/g) | SF\(_{Cs/Sr}\) |
|--------------------------|-----------------|-----------------|-----------------|-----------------|---------------|---------------|----------------|----------------|-------------|
| 0.03                     | 4.839           | 4.7985          | 237             | 142             | 40.08         | 0.84          | 0.67 \(\times 10^3\) | 8.44           | 79.38       |
| 0.11                     | 6.34            | 5.58            | 87              | 11              | 87.36         | 11.99         | 6.91 \(\times 10^3\) | 136.2          | 50.73       |
| 1.86                     | 5.36            | 5.144           | 4.36            | 1.385           | 68.23         | 4.03          | 2.15 \(\times 10^3\) | 41.99          | 51.16       |
| 17.17                    | 49.364          | 46.516          | 4.285           | 1.33            | 68.96         | 5.77          | 2.22 \(\times 10^3\) | 61.23          | 36.26       |
| 75.55                    | 254             | 242             | 5.1             | 1.7             | 66.67         | 4.72          | 2.0 \(\times 10^3\)  | 49.59          | 40.33       |

Table S16. The results for the Cs\(^+\)-exchange of 1 and 2 in the competitive experiments with mixed metal ions (V/m = 1000 mL/g, room temperature, 10 h contact time).

| Compounds | C\(_0\) (mg/L) | C\(_e\) (mg/L) | R (%) | K\(_d\) (mL/g) |
|-----------|----------------|----------------|-------|---------------|
| 1         | C\(_0\)Cs (Cs) | C\(_e\)Cs (Cs) | R\(_{Cs}\) (Cs) | 6.08 \(\times 10^3\) (Cs) |
|           | 0.9045         | 85.87          | 7.22 \(\times 10^3\) |
|           | 38.84 (K)      | 42.05          | 19.82 (Na)     |
|           | 54.5 (Na)      | 1.94 (Na)      | 54.60 (Na)     |
|           | 43.60 (Ca)     | 1.69 (Ca)      | 43.86 (Ca)     |
|           | 46.92 (Mg)     | 3.68 (Mg)      | 47.61 (Mg)     |
|           | 59.725 (Sr)    | 3.37 (Sr)      | 58.755 (Sr)    |
| 2         | C\(_0\)Cs (Cs) | C\(_e\)Cs (Cs) | R\(_{Cs}\) (Cs) | 2.10 \(\times 10^3\) (Cs) |
|           | 2.065          | 67.73 (Cs)     | 9.12 \(\times 10^3\) |
|           | 34.24 (K)      | 48.91 (K)      | 96.96 (K)      |
|           | 54.60 (Na)     | 1.76 (Na)      | 17.95 (Na)     |
|           | 43.86 (Ca)     | 1.10 (Ca)      | 11.17 (Ca)     |
|           | 47.61 (Mg)     | 2.28 (Mg)      | 23.31 (Mg)     |
|           | 58.755 (Sr)    | 4.94 (Sr)      | 52 (Sr)        |

Table S17. The results of Cs\(^+\)-adsorption for 1 in tap water, Minjiang water, river water, simulated groundwater and sea water, containing low concentration of Cs\(^+\) (V = 10 mL, m = 10 mg, V/m = 1000 mL/g; 10 h contact time; at room temperature).

| Experimental condition | C\(_e\)Cs (mg/L) | K\(_d\)Cs (mL/g) |
|------------------------|-----------------|-----------------|
| Tap water, 7.40 mg/L Cs\(^+\) + 5.03 mg/L K\(^+\) + 21.85 mg/L Na\(^+\) + 12.26 mg/L Ca\(^{2+}\) + 2.64 mg/L Mg\(^{2+}\) | 0.90            | 7.22 \(\times 10^3\) |
| Minjiang water, 5.31 mg/L Cs\(^+\) + 3.725 mg/L K\(^+\) + 17.108 mg/L Na\(^+\) + 10.242 mg/L Ca\(^{2+}\) + 1.967 mg/L Mg\(^{2+}\) | 0.57            | 8.32 \(\times 10^3\) |
| River water, 8.955 mg/L Cs\(^+\) + 1.987 mg/L K\(^+\) + 2.664 mg/L Na\(^+\) + 3.061 mg/L Ca\(^{2+}\) + 0.431 mg/L Mg\(^{2+}\) | 0.885           | 9.12 \(\times 10^3\) |
| Simulated groundwater, 4.575 mg/L Cs\(^+\) + 9.82 mg/L K\(^+\) + 375.5 mg/L Na\(^+\) + 9.38 mg/L Ca\(^{2+}\) + 4.945 mg/L Mg\(^{2+}\) | 0.595           | 6.69 \(\times 10^3\) |
| Sea water, 9.725 mg/L Cs\(^+\) + 327.05 mg/L K\(^+\) + 9546 mg/L Na\(^+\) + 366.1 mg/L Ca\(^{2+}\) + 1081.5 mg/L Mg\(^{2+}\) | 9.565           | 16.73            |
Table S18. The results of Cs⁺-adsorption for 2 in tap water, Minjiang water, river water, simulated groundwater and sea water, containing low concentration of Cs⁺ ($V = 10$ mL, $m = 10$ mg, $V/m = 1000$ mL/g; 10 h contact time; at room temperature).

| Experimental condition | $C_0^{Cs}$ (mg/L) | $R^{Cs}$ (%) | $K_d^{Cs}$ (mL/g) |
|------------------------|-------------------|---------------|-------------------|
| Tap water, 7.40 mg/L Cs⁺ + 5.03 mg/L K⁺ + 21.85 mg/L Na⁺ + 12.26 mg/L Ca²⁺ + 2.64 mg/L Mg²⁺ | 0.90 | 87.84 | 7.22 × 10³ |
| Minjiang water, 5.8 mg/L Cs⁺ + 3.69 mg/L K⁺ + 16.72 mg/L Na⁺ + 11.02 mg/L Ca²⁺ + 1.921 mg/L Mg²⁺ | 0.7125 | 87.72 | 7.14 × 10³ |
| River water, 8.935 mg/L Cs⁺ + 2.77 mg/L K⁺ + 4.52 mg/L Na⁺ + 6.71 mg/L Ca²⁺ + 0.98 mg/L Mg²⁺ | 1.77 | 80.19 | 4.04 × 10³ |
| Simulated groundwater, 4.575 mg/L Cs⁺ + 9.82 mg/L K⁺ + 375.5 mg/L Na⁺ + 9.38 mg/L Ca²⁺ + 4.945 mg/L Mg²⁺ | 1.325 | 71.04 | 2.45 × 10³ |
| Sea water, 8.39 mg/L Cs⁺ + 310.5 mg/L K⁺ + 8645 mg/L Na⁺ + 369.5 mg/L Ca²⁺ + 988.5 mg/L Mg²⁺ | 8.32 | 0.83 | 8.41 |

Section S6. γ radiation resistances studies

Figure S13. (a) PXRD patterns for simulated and as-synthesized 1 and samples after γ irradiation; (b) PXRD patterns for simulated and as-synthesized 2 and samples after γ irradiation.

Table S19. Data for Cs⁺ ion exchange of 1 and 2 before and after irradiation.

| Samples          | 1                               | 2                               |
|------------------|---------------------------------|---------------------------------|
|                  | Pristine | 100 kGy γ irradiation | 200 kGy γ irradiation | Pristine | 100 kGy γ irradiation | 200 kGy γ irradiation |
| $C_0^{Cs}$ (mg/L) | 21.4     | 21.4               | 21.4              | 21.4     | 21.4               | 21.4              |
| $C_e^{Cs}$ (mg/L) | 2.255    | 2.065              | 2.46              | 3.555    | 3.365              | 3.485             |
| $R^{Cs}$ (%)      | 89.46    | 90.35              | 88.50             | 83.39    | 84.28              | 83.71             |
| $K_d^{Cs}$ (mL/g) | 8.49 × 10³ | 9.36 × 10³          | 7.70 × 10³        | 5.02 × 10³ | 5.36 × 10³          | 5.14 × 10³        |
| $C_0^{Cs}$ (mg/L) | 40.675   | 40.675             | 40.675            | 40.675   | 40.675             | 40.675            |
| $C_e^{Cs}$ (mg/L) | 3.8      | 3.65               | 4.15              | 10.475   | 11.075             | 11.775            |
Section S7. Elution and Recycle

| $R^{Cs}_t$ (%) | 90.66 | 91.03 | 89.80 | 74.25 | 72.77 | 71.05 |
|----------------|-------|-------|-------|-------|-------|-------|
| $K_d^{Cs}$ (mL/g) | $9.70 \times 10^3$ | $1.01 \times 10^4$ | $8.80 \times 10^3$ | $2.88 \times 10^3$ | $2.67 \times 10^3$ | $2.45 \times 10^3$ |

**Figure S14.** EDS analysis results of the eluted products for Cs$^+$-exchanged 1 and 2 by using the 0.3 M KCl solution.

**Figure S15.** SEM images and elemental distribution maps of In and K of (a) 1-K and (b) elution product of 2-Cs, respectively. (c) X-ray photoelectron survey spectra for 1 and 1-K. (d) X-ray photoelectron spectrum of potassium for 1-K. EDS analysis results for the
Table S20. Data for the cycle of 1-K.

| Cycle   | $C_0^{Cs}$ (mg/L) | $C_{e}^{Cs}$ (mg/L) | $R^{Cs}$ (%) |
|---------|------------------|-------------------|--------------|
| Cycle 1 | 8.735            | 1.32              | 84.89        |
|         | 17.4             | 2.23              | 87.18        |
| Cycle 2 | 8.735            | 1.26              | 85.58        |
|         | 17.4             | 2.19              | 87.41        |
| Cycle 3 | 8.735            | 1.705             | 80.48        |
|         | 17.4             | 3.655             | 78.99        |

Figure S16. (a) PXRD patterns for 1, 1-Cs and the eluted product 1-K in cycle 0. PXRD patterns of 1-Cs and 1-Cs-K after first (b), second (c) and third (d) adsorption-elution cycles. The subscripts of 1-Cs indicate different ion exchange experiments with solutions containing various initial Cs$^+$ ions concentrations (1-Cs$_1$: 8.735 mg/L and 1-Cs$_2$: 17.4 mg/L). 1-Cs$_1$-K and 1-Cs$_2$-K indicate the eluted products of 1-Cs$_1$ and 1-Cs$_2$ by using the 0.3 M KCl solution, respectively. EDS analysis results for the 1-Cs$_1$-K (e) and 1-Cs$_2$-K (f) in cycle 3.