

Supplementary Material

# Top-Down Synthesis of NaP Zeolite from Natural Zeolite for the Higher Removal Efficiency of Cs, Sr, and Ni

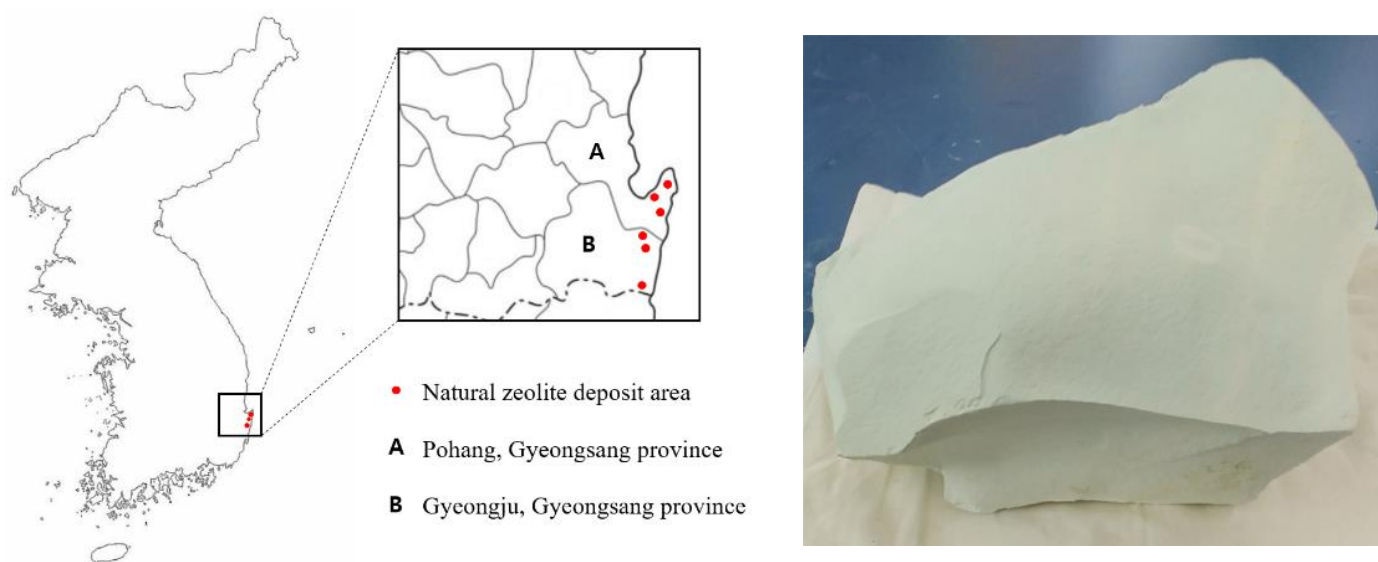
Seokju Hong <sup>1</sup> and Wooyong Um <sup>1,2,3,\*</sup>

<sup>1</sup> Division of Advanced Nuclear Engineering (DANE), Pohang University of Science and Technology (POSTECH), 77 Chongam-ro, Nam-Gu, Pohang 790-784, Korea; frederic@postech.ac.kr

<sup>2</sup> Division of Environmental Sciences and Engineering (DESE), Pohang University of Science and Technology (POSTECH), 77 Chongam-ro, Nam-Gu, Pohang 790-784, Korea

<sup>3</sup> Nuclear Environmental Technology Institute (NETI), Pohang University of Science and Technology (POSTECH), Pohang, Gyeongbuk 790-784, Korea

\* Correspondence: wooyongum@postech.ac.kr; Phone: +82-54-279-9563; Fax: +82-54-279-9559



**Figure S1.** The location of a natural Korean zeolite-mining site (left) where the natural zeolite ore sample (right) was collected.

**Table S1.** Composition of Hanbit groundwater.

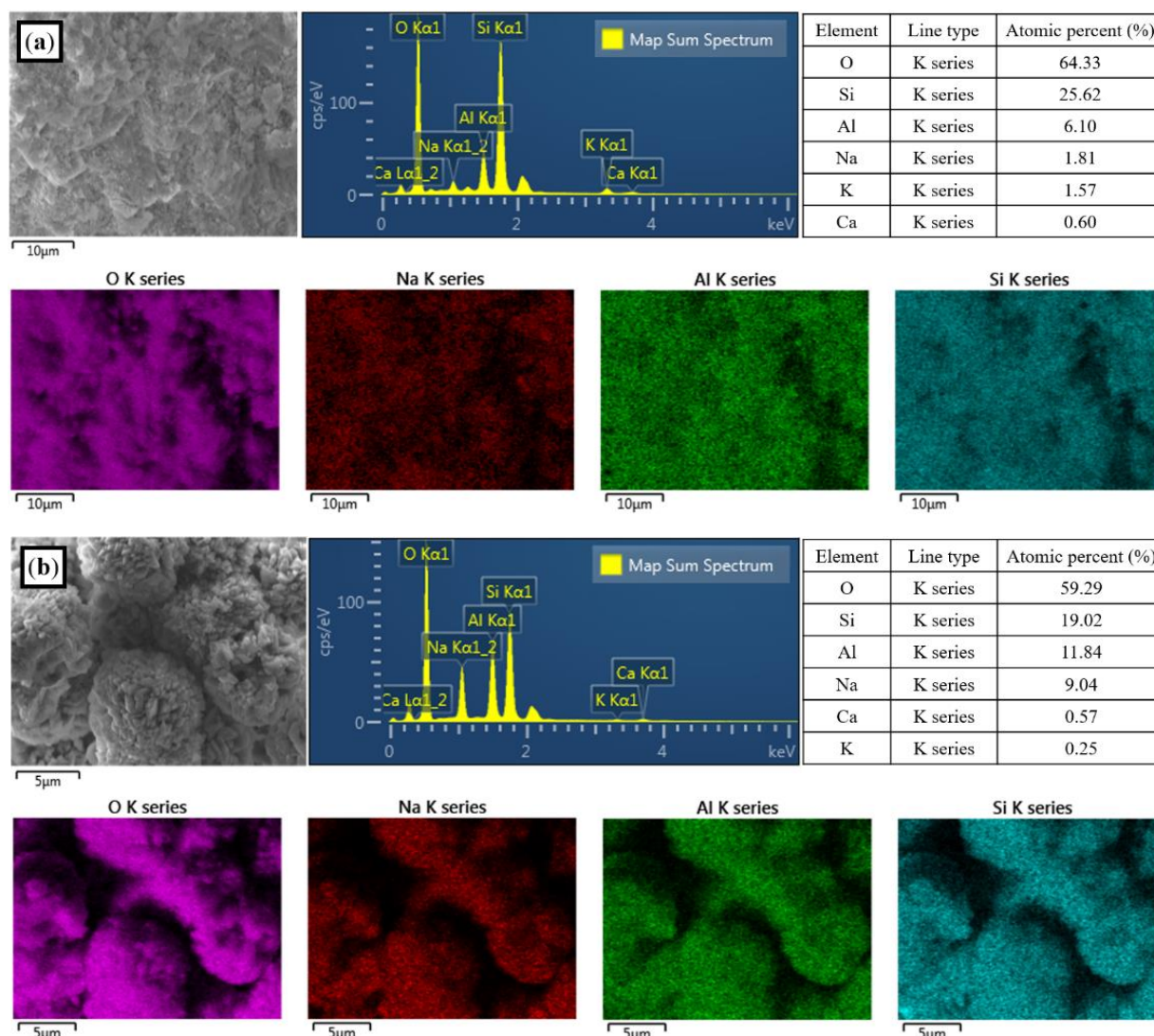
Composition	Groundwater (ppm)
Na <sup>+</sup>	45.9
K <sup>+</sup>	2.45
Mg <sup>2+</sup>	9.76
Ca <sup>2+</sup>	54.5
Cl <sup>-</sup>	28.0
SO <sub>4</sub> <sup>2-</sup>	0.81
NO <sub>3</sub> <sup>-</sup>	9.4



**Figure S2.** The “top-down” synthesis approach to produce granular-sized zeolites: (a) natural zeolite ore, (b) crushed natural zeolite (granular-sized), (c) zeolite immersed in NaOH solution, and (d) during the reaction process in the oven, and (e) resulting as-synthesized NaP zeolite (granular-sized).

**Table S2.** Chemical compositions of granular-sized natural and NaP zeolites expressed in wt.%.

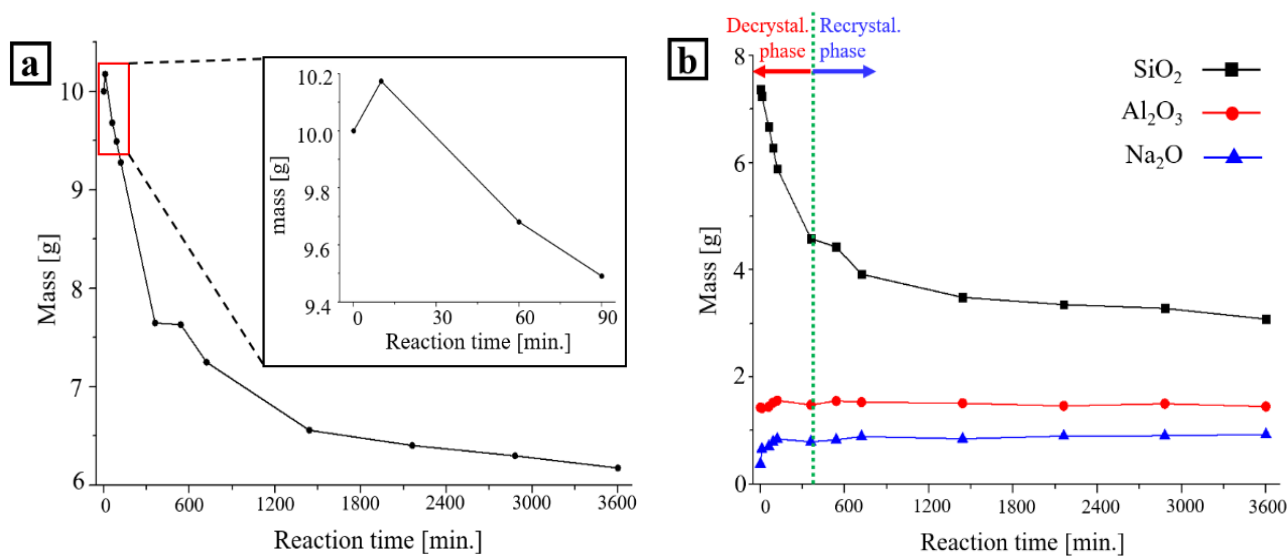
Compound	Natural Zeolite	NaP Zeolite
SiO <sub>2</sub>	72.43	48.70
Al <sub>2</sub> O <sub>3</sub>	14.10	23.61
Na <sub>2</sub> O	3.51	14.8
K <sub>2</sub> O	2.36	1.17
CaO	2.08	3.41
MgO	0.81	1.37
Others	3.81	6.94



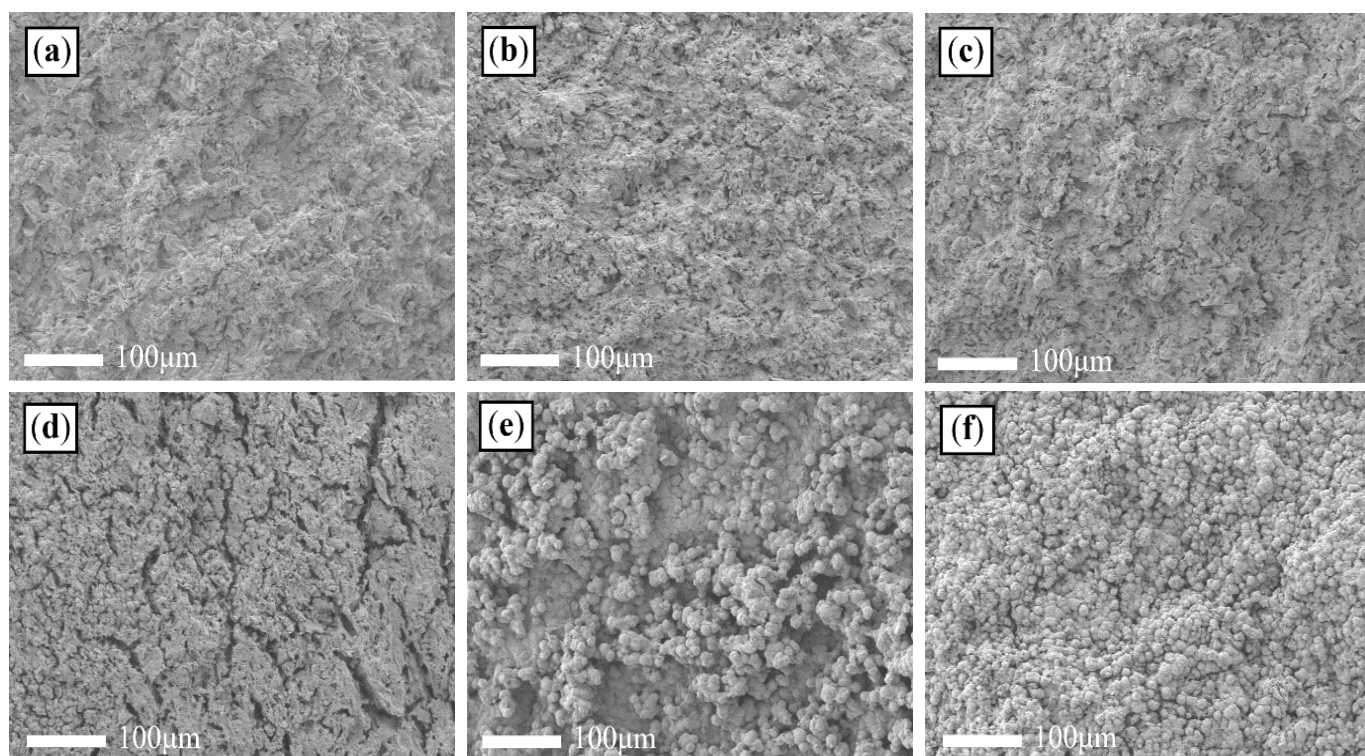
**Figure S3.** Elemental mapping of natural zeolite (a) and NaP zeolite (b) with SEM-EDX analysis.

**Table S3.** Textural parameters of natural and NaP zeolites.

	Natural Zeolite (powder-sized)	NaP Zeolite (powder-sized)	Natural Zeolite (granular-sized)	NaP Zeolite (granular-sized)
Specific surface area ( $S_{\text{BET}}$ ) ( $\text{m}^2/\text{g}$ )	34.446	89.298	31.349	95.949
Total pore volume ( $\text{cm}^3/\text{g}$ )	0.122	0.139	0.077	0.099
Micropore volume ( $\text{cm}^3/\text{g}$ )	0.016	0.036	0.016	0.021

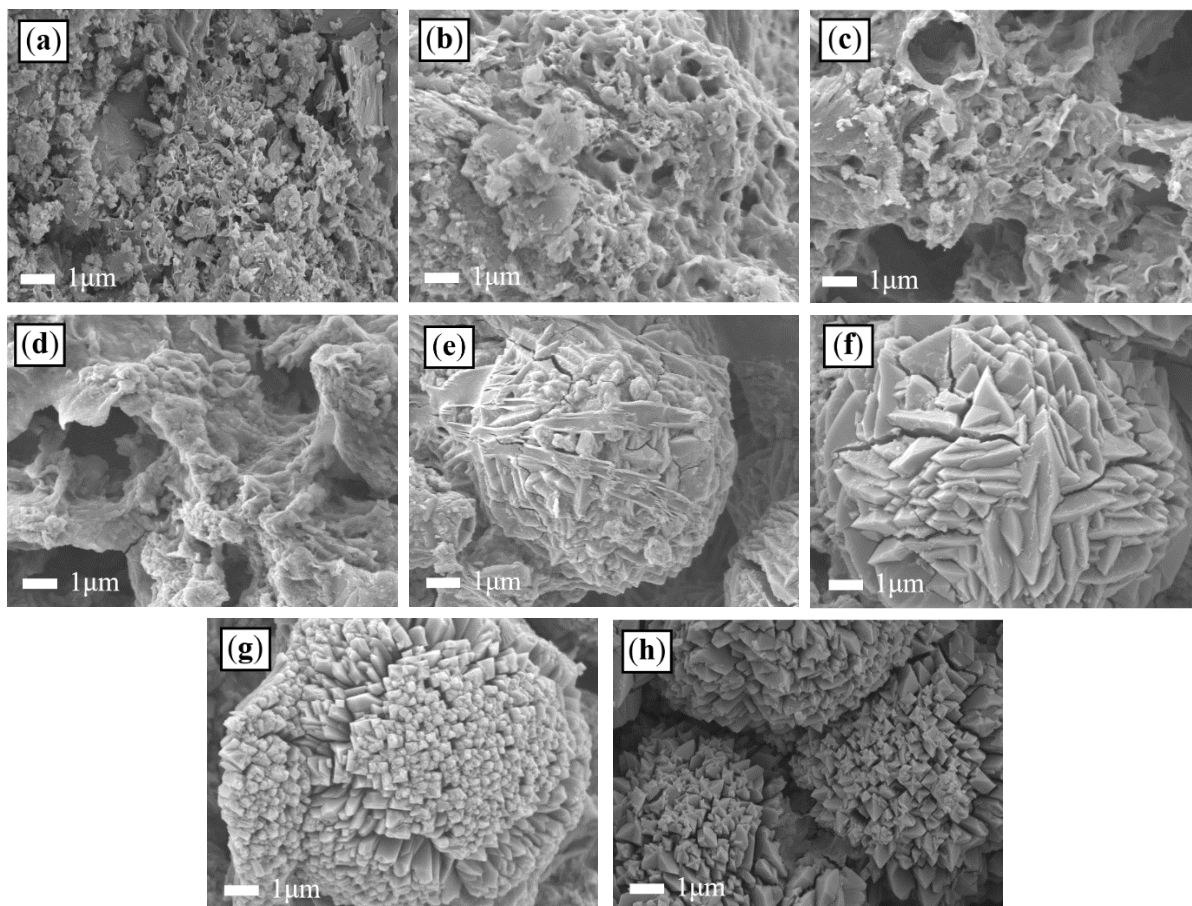


**Figure S4.** Mass variation (a) and composition variation (b) of zeolite with different reaction times.

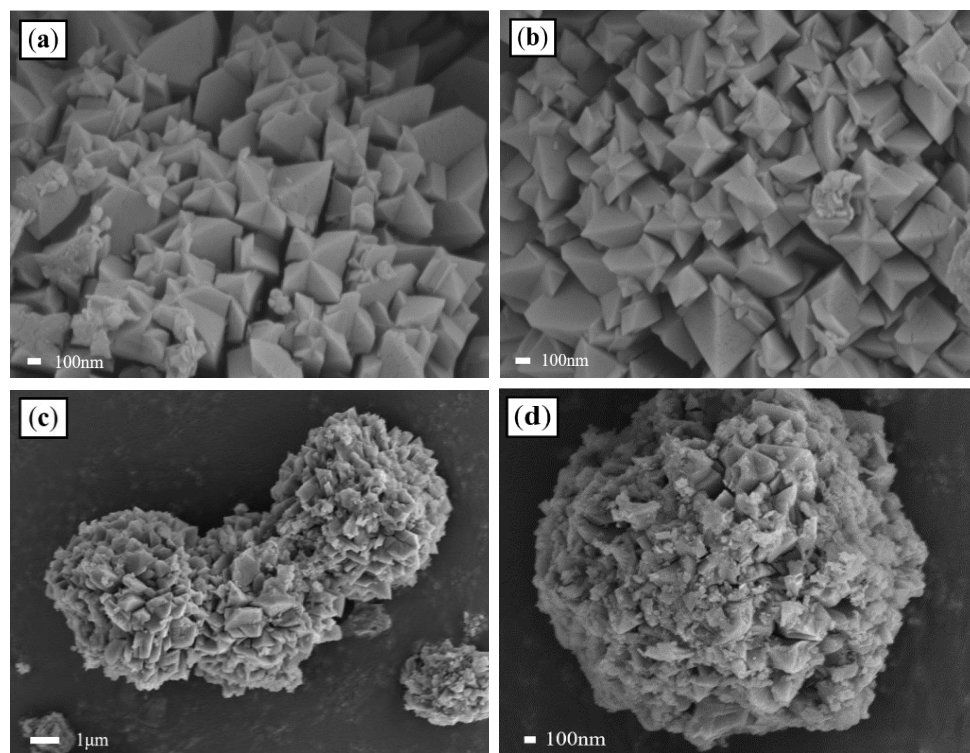


**Figure S5.** SEM images showing morphological changes at different reaction times; (a) 10 min, (b) 1 h, (c) 2 h, (d) 9 h, (e), 24 h, and (f) 48 h.

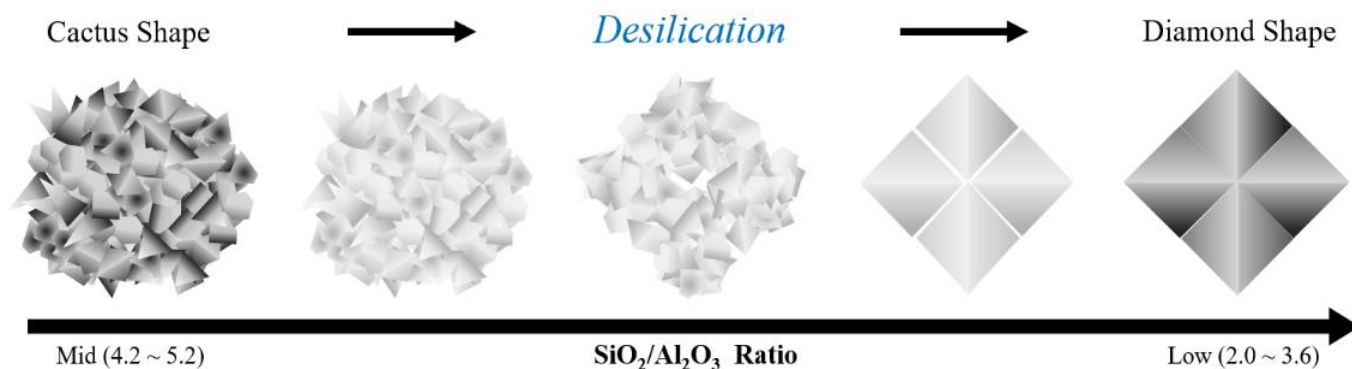




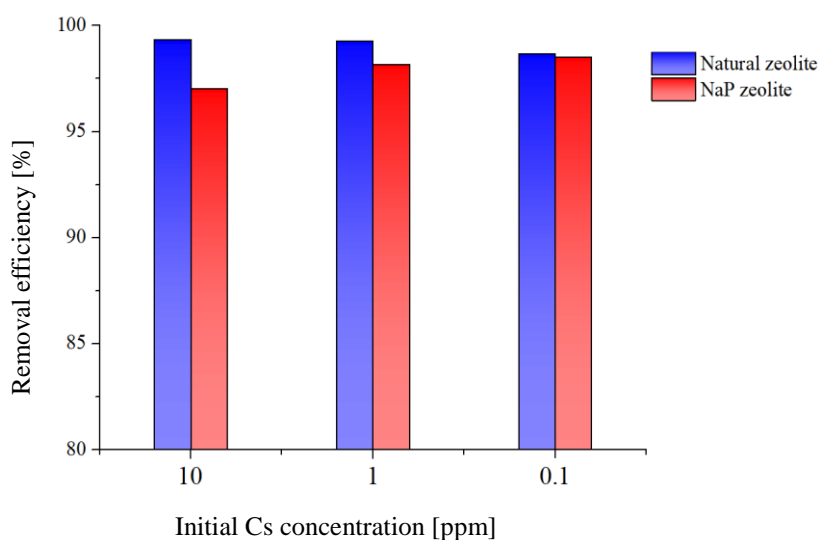
**Figure S6.** SEM images showing morphological changes at different reaction times; (a) Natural zeolite, (b) 10 min, (c) 1 h, (d) 2 h, (e) 9 h, (f) 24 h, (g) 48 h, and (h) 60 h.



**Figure 7.** SEM images of two different-sized NaP zeolites (a), (b): Granular-sized NaP zeolite's surface, (c), (d): Powder-sized NaP zeolite's surface.



**Figure S8.** Morphological transition of the NaP zeolite with different Si/Al ratios.



**Figure S9.** Batch sorption measurements for the removal of Cs at various concentrations in groundwater collected around the Hanbit NPP site. (Note: Error bars were omitted because of size constraints. The initial  $\text{Cs}^+$  concentrations were set to 10, 1, and 0.1 ppm, with the solid-to-solution ratio at 100 mL/g.