SUPPLEMENTARY INFORMATION

Which Type of Clay Minerals Fix Cesium Ion Effectively? -Cavity-Charge Matching Effect-

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Ion exchange of clay minerals by NaTPB  (Artificial weathering)

To uniform the weathering degree, an ion exchange from $\text{K}^+$ to $\text{Na}^+$ by NaTPB was carried out for vermiculite, phlogopite G325, phlogopite N and biotite, according to the procedure shown in experimental section. Since $\text{Na}^+$ is the original counter cation for Saponite, the ion exchange is not necessary. The time course of replacement ratio from $\text{K}^+$ to $\text{Na}^+$ is shown in Figure S1. By the artificial weathering treatment for 1000hrs, the replacement ratio reached up to 97.6, 87.7, 90.7, 86.6 % for Vermiculite, Phlogopite G325, Phlogopite N and Biotite, respectively.

XRD patterns were examined for the $\text{Na}^+$ substituted clay minerals. The patterns for Phlogopite G325 before and after NaTPB treatment as the typical cases are shown in Figure S2. While untreated $\text{K}^+$- Phlogopite G325 exhibits clear peak at 8.82˚ (Fig. S2 (i)), $\text{Na}^+$ substituted one shows peak at 7.28˚ (Fig. S2 (ii)) under the ambient conditions. These observations indicate that interlayer distance ($d$) was enlarged from 0.07 nm to 0.26 nm by the $\text{Na}^+$ substitution. This expansion can be explained by the hydration
behavior of Na⁺. In fact, the peak was shifted to 6.0° (d = 0.52 nm) and 9.1° (d = 0.01 nm) under the wet and dry condition, respectively. These changes were reversible. On the other hand, XRD pattern for K⁺- Phlogopite G325 did not show any dependence on the humidity change.

![Figure S1](image_url)

**Figure S1.** Time-course of substitution ratio from K⁺ to Na⁺ for vermiculite, phlogopite G325, phlogopite N and biotite.
Figure S2. XRD patterns for K$^+$ (i) and Na$^+$ (ii) - Phlogopite G325. The patterns for swelled (iii) and unhydrated condition (iv) are shown for Na$^+$ - Phlogopite G325.
Figure S3. Time-course for Cs\textsuperscript+ adsorption by Na\textsuperscript+ substituted clay minerals.
Figure S4. XRD pattern for Phlogopite GS325 (top) and Phlogopite N (bottom) under dry (broken line) and wet (solid line) conditions.
Figure S5. Two clay sheets, where \((\text{number of negative charge} /\text{number of SDC cavity of the clay}) = 0.5\), can form interlocking structure.
Figure S6. The relationship between (number of negative charge / number of Siloxane Ditrigonal Cavity of the clay) and the desorption ratio. Open symbols and dotted line: desorption behavior, closed symbols and bold line: adsorption behavior.
Table S1. Number of negative charge and SDC per unit

<table>
<thead>
<tr>
<th>Clay</th>
<th>Number of negative charge per unit (a)</th>
<th>Number of SDC per unit (b)</th>
<th>(a)/(b)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Saponite</td>
<td>0.4</td>
<td>2</td>
<td>0.20</td>
</tr>
<tr>
<td>Vermiculite</td>
<td>0.9</td>
<td>2</td>
<td>0.45</td>
</tr>
<tr>
<td>Phlogopite G325</td>
<td>1.0</td>
<td>2</td>
<td>0.50</td>
</tr>
<tr>
<td>Phlogopite N</td>
<td>1.2</td>
<td>2</td>
<td>0.60</td>
</tr>
<tr>
<td>Biotite</td>
<td>1.3</td>
<td>2</td>
<td>0.65</td>
</tr>
</tbody>
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