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 K^+ to Na^+ by NaTPB was carried out for vermiculite, phlogopite G325, phlogopite N and biotite, according to the procedure shown in experimental section. Since Na^+ is the original counter cation for Saponite, the ion exchange is not necessary. The time course of replacement ratio from K^+ to 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 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 K^+- Phlogopite G325 exhibits clear peak at 8.82˚ (Fig. S2 (i)), 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 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.

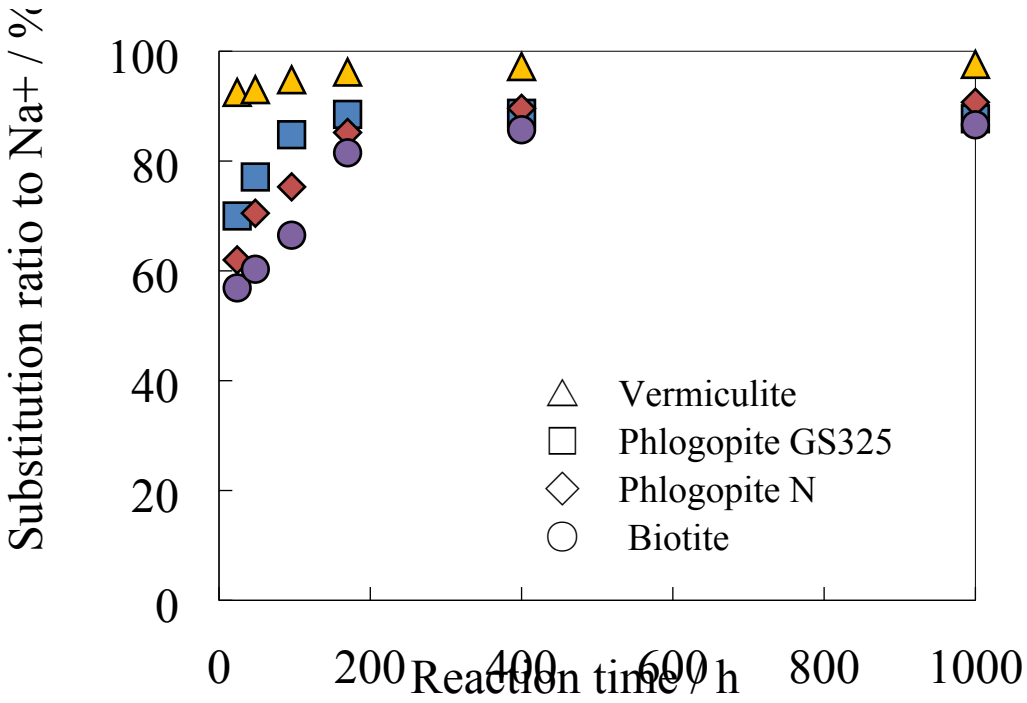

Figure S1. Time-course of substitution ratio from K^+ to Na^+ for vermiculite, phlogopite G325, phologopite N and biotite.

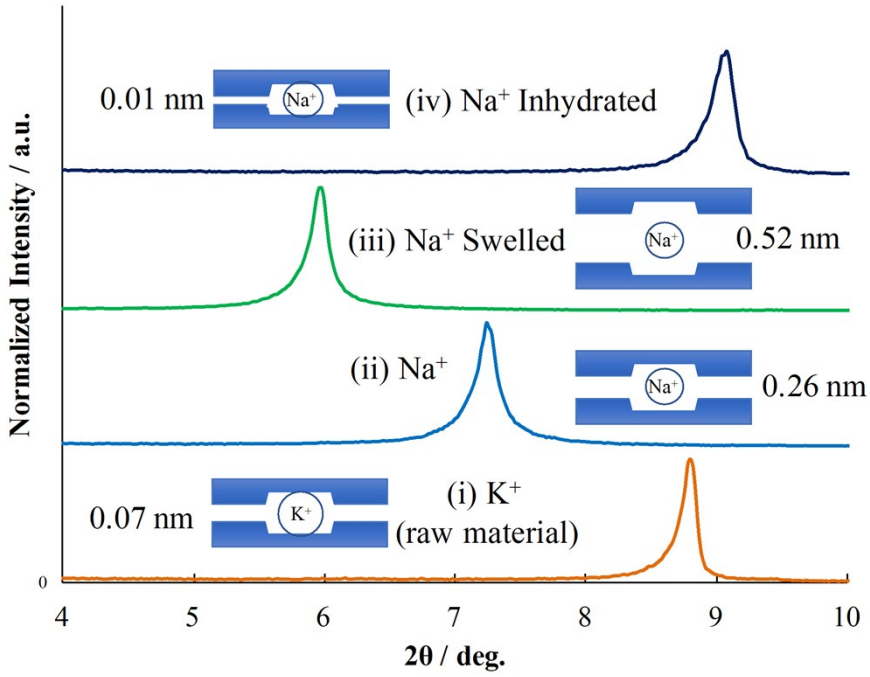

Figure S2. XRD patterns for K^+ (i) and Na^+ (ii) - Phlogopite G325. The patterns for swelled (iii) and unhydrated condition (iv) are shown for Na^+ - Phlogopite G325.

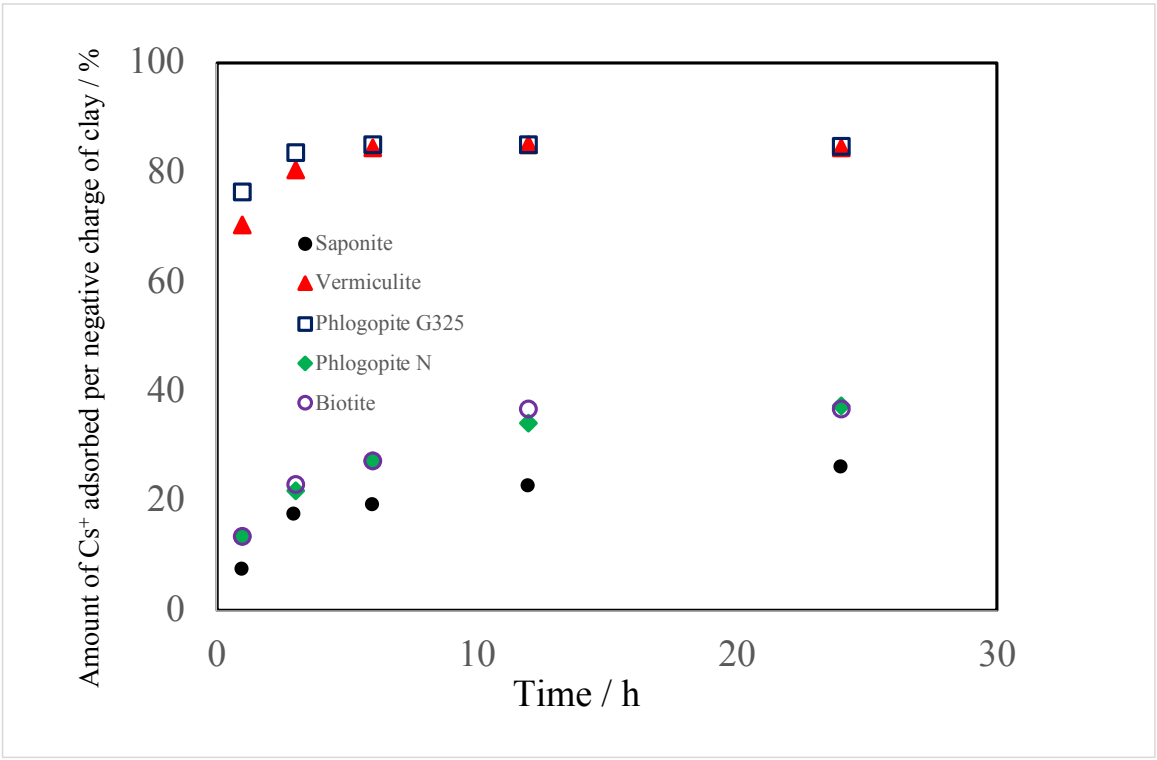

Figure S3. Time-course for Cs^+ adsorption by Na^+ substituted clay minerals.

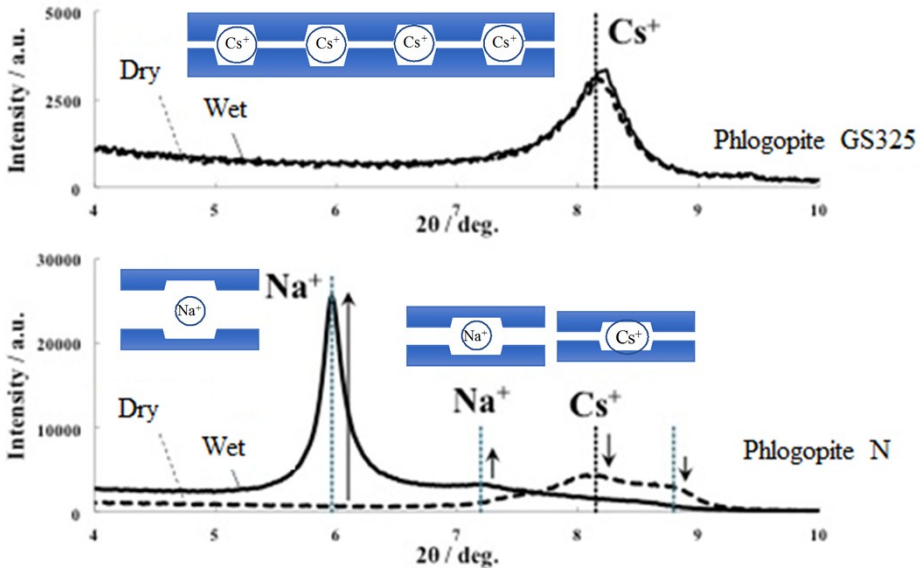

Figure S4. XRD pattern for Phlogopite GS325 (top) and Phlogopite N (bottom) under dry (broken line) and wet (solid line) conditions.

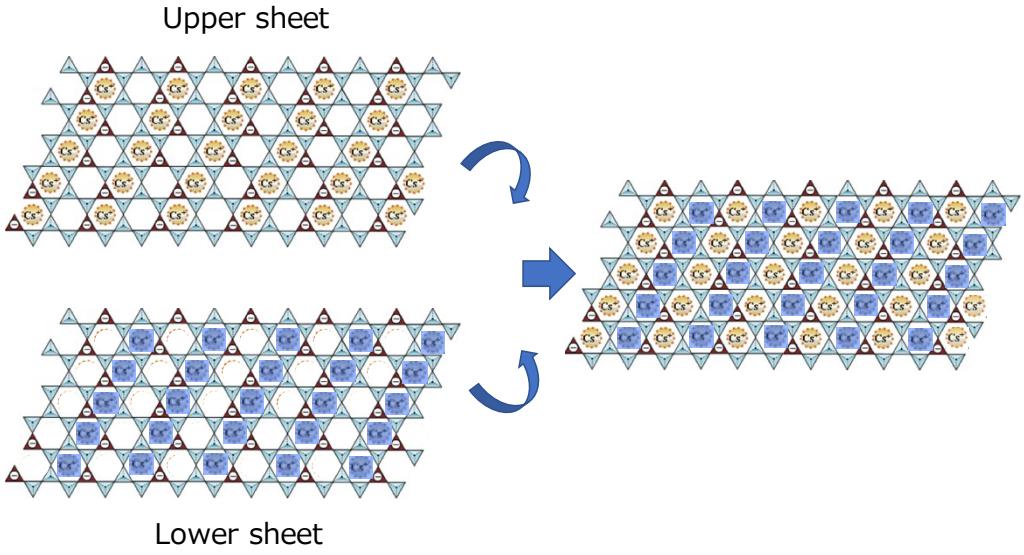

Figure S5. Two clay sheets, where (number of negative charge / number of SDC cavity of the clay) = 0.5, can form interlocking structure.

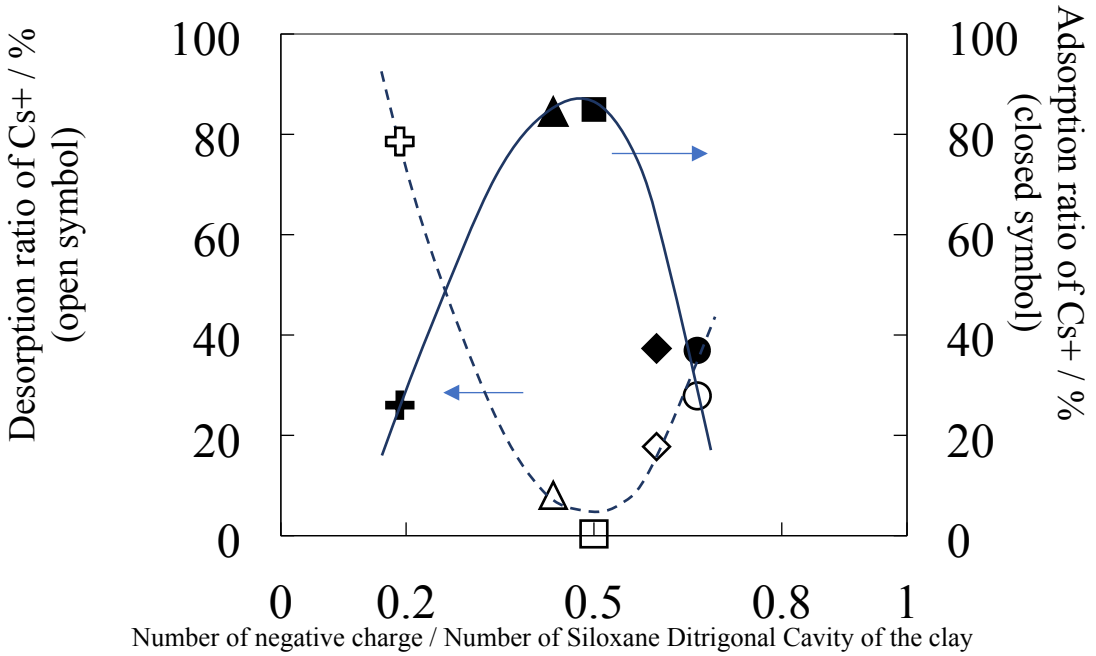

Figure S6. The relationship between (number of negative charge / number of SDC cavity of the clay) and the desorption ratio. Open symbols and dotted line: desorption behavior, closed symbols and bold line: adsorption behavior.

Table S1. Number of negative charge and SDC per unit

Clay	Number of negative charge per unit (a)	Number of SDC per unit (b)	(a)/(b)
Saponite	0.4	2	0.20
Vermiculite	0.9	2	0.45
Phlogopite G325	1.0	2	0.50
Phlogopite N	1.2	2	0.60
Biotite	1.3	2	0.65