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Extraction of Cs bound with biotite by addition of oxalic acid without heating

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27 **Abstract**

28 **Purpose**

29 There is a large quantity of contaminated soil with radionuclides due to the accident of Fukushima Daiichi
30 Nuclear Power Plant in Japan. Some previous studies reported that Cs^+ could be desorbed from soil with
31 high efficiency, however, these methods required huge input energy for operating. Therefore, we focused
32 on oxalic acid, which has shown relatively high desorption efficiency in previous thermal treatment, and
33 evaluated the potential of oxalic acid as a leaching reagent to reduce the volume of contaminated soil
34 without heating.

35 **Methods**

36 Stable isotope Cs^+ contaminated biotite was retained in plastic bag for 3 months for aging. Oxalic acid was
37 added to the contaminated biotite and the mixture was maintained around 20 °C. After each leaching time,
38 the mixture was separated using filter paper and the concentration of Cs^+ in filtrate was measured. Structural
39 changes in biotite before and after leaching were also analyzed. LiNO_3 was also used for comparison studies.

40 **Results**

41 The desorption efficiency of Cs^+ from biotite by leaching with oxalic acid was 94.9% after 2 weeks and the
42 intensity of the specific reflection for biotite in the X-ray diffractograms decreased with leaching time.
43 These results indicate that oxalic acid can remove Cs^+ from biotite through the decomposition of the biotite
44 structure. Since LiNO_3 desorbed only 32.2% of Cs^+ as ion-exchangeable form after 2 weeks, oxalic acid

45 can desorb Cs^+ , which has a strong interaction with biotite such as the inner-sphere complex, without
46 heating.

47 **Conclusion**

48 Although the desorption of Cs^+ from biotite by leaching with oxalic acid leaching takes a long time, it is a
49 great advantage that this leaching method does not require external energy such as heating. Therefore,
50 oxalic acid leaching has high potential to reduce the volume of contaminated soil with radionuclides.

51

52 **Keywords:** Cesium desorption; Biotite; Oxalic acid leaching; Decomposition; Without heating; Volume
53 reduction

54

55

56 **1 Introduction**

57 The accident at the Fukushima Daiichi Nuclear Power Plant happened in Japan on March 2011. The
58 quantity of contaminated soil was estimated to be 13-14 million m³, of which the clayey soil discharged
59 from agricultural fields and forests was estimated to be 6 million m³ (Japan Ministry of the Environment,
60 2019). The transportation of these soils from the temporary storage site to the interim storage facility started
61 in 2015 (Japan Ministry of the Environment, 2018). It is estimated that even after a ¹³⁷Cs half-life of 30
62 years, the volume of soil above 8,000 Bq/kg will be 105 thousand m³, of which 98 thousand m³ is clayey
63 soil (Japan Ministry of the Environment, 2018). The contaminated soils with less than 8,000 Bq/kg are
64 intended to be recycled as inorganic civil engineering material, while the contaminated soils with more than
65 8,000 Bq/kg are difficult to utilize as resources. In Japan where there are many natural disasters, the safe
66 storage of the large quantity of contaminated soil for a long time is limited. Therefore, the removal of
67 radioactive Cs from contaminated soil is urgently needed to reduce the volume of contaminated soil.
68 However, Cs⁺ forms an inner-sphere complex, which directly coordinates without the intervention of water
69 molecules, with the interlayers of clay minerals, and this interaction is too strong to separate Cs⁺ from clay
70 minerals (Mukai et al. 2018). Recently, low-pressure sublimation, thermal, and microwave treatments have
71 been developed to desorb Cs⁺ from the contaminated soil (Shimoyama et al. 2014; Parajuli et al. 2015; Yin
72 et al. 2017; Tamura et al. 2021; Akemoto et al. 2021a). These methods have high desorption efficiency of
73 Cs⁺ from soil in relatively short time, however, heating at the boiling point of water or more is required.

74 Therefore, these method have limits to treat with large quantity of contaminated soil.

75 In this study, we focused on oxalic acid which has relatively high desorption efficiency in previous thermal
76 treatment without a special experimental environment (Kim et al. 2020). The potential of oxalic acid was
77 evaluated by the desorption experiment of Cs^+ from model contaminated biotite without heating.

78

79 **2 Materials and methods**

80 **2.1 Clay mineral and reagents**

81 Biotite in Tamura-gun, Fukushima Prefecture, Japan (Geo-Science Materials Nichika Inc., Japan) was used
82 as the model clay mineral. Since the adsorption behavior of Cs^+ on this biotite was established by the
83 Freundlich adsorption isotherm, the interlayer and surface of biotite acted as adsorption sites for Cs^+
84 (Akemoto et al. 2021b). For characteristics of biotite used in this study, cation exchange capacity is 380
85 mmol/kg and chemical component is shown in **Table S1**. From the K_2O content in the biotite, the biotite
86 used in our study is suggested to be more weathering and then it may be difficult to desorb Cs^+ from the
87 biotite by the ordinal condition (Kitayama et al. 2020). Cesium chloride (guaranteed reagent grade, >99%)
88 and oxalic acid (guaranteed reagent grade, 98%) were purchased from Fujifilm Wako Pure Chemical
89 Corporation, Japan. Lithium nitrate (guaranteed reagent grade, >98%) was purchased from Kanto Chemical
90 Co., Inc., Japan. All reagents were used without pretreatment.

91

92 2.2 Desorption experiment

93 The Cs⁺ contaminated biotite was prepared as described in a previous study (Akemoto et al. 2021b). The
94 particle size of biotite was less than 53 μm and the adsorption capacity of biotite for Cs⁺ was 31.9 mg/g.
95 Biotite that adsorbed Cs⁺ was retained in plastic bag at room temperature for 3 months before experiments.
96 Oxalic acid (0.5 mol/L, 25 mL) was added to the contaminated biotite (0.05 g) and the mixture was
97 maintained without any mechanical shaking under room temperature, which was measured continuously
98 during the experiment (Thermo Recorder, TR-71U, T&D Corporation, Japan). After each leaching time (1
99 hour, 1 day, 3 days, 1 week, 2 weeks, 3 weeks, and 4 weeks), the mixture was separated using a filter paper
100 (5C, 1 μm pore size, Toyo Roshi Kaisha Ltd., Japan), and the concentration of Cs⁺ in the filtrate was
101 measured using a flame atomic absorption spectrophotometer (FAAS, ZA-3300, Hitachi Ltd., Japan) at
102 852.1 nm. Oxalic acid at low concentration (0.1 mol/L) and LiNO₃ (1.0 mol/L) were also used for
103 comparison studies for their effect on extraction of Cs⁺ with the same methodology as mentioned above.
104 Since Li⁺ has a high hydration energy, it is expected to desorb Cs⁺ by expansion of the clay mineral
105 interlayer due to the penetration of Li⁺ in it (Mukai et al. 2018). The desorption efficiency of Cs⁺ from
106 biotite was calculated by the following equation (Eq. 1).

$$107 \quad \text{Desorption efficiency (\%)} = \frac{C_t \times V}{Q \times W} \times 100 \quad (1)$$

108 where C_t is the concentration of Cs⁺ (mg/L) in the solution after each leaching time t , V is the volume of
109 the extract solution (L), Q is the adsorption capacity of biotite for Cs⁺ (31.9 mg/g), and W is the amount of

110 biotite (g). All experiments were performed in triplicates. The main elements of the clay minerals (Al, Fe,
111 Mg, and Si) in the filtrate were measured using inductively coupled plasma-atomic emission spectroscopy
112 (ICP-AES, ICPS-8100, Shimadzu Corporation, Japan). The percentage of these elements (Al, Fe, Mg, and
113 Si) extracted from the initial biotite was calculated based on the results of energy-dispersive X-ray
114 fluorescence spectrometry (XRF, JSX-3100RII, JEOL Ltd., Japan) (Akemoto et al. 2021b). The solution
115 pH was measured using a pH meter (D-51, Horiba Ltd., Japan). The biotite before and after leaching
116 treatment was measured using powder X-ray diffraction (XRD, SmartLab, Rigaku Corporation, Japan).
117 XRD was performed using Cu K α radiation ($\lambda = 0.154$ nm) at 40 kV and 30 mA at a speed of 1 °/min and
118 an angular step of 0.01°.

119

120 **3 Results and discussion**

121 The room temperature was observed to be at 20 ± 5 °C during the 4 weeks experiment. The pH of the
122 solutions were 5.9, 1.3 and 0.8 in 1.0 mol/L LiNO₃, 0.1 mol/L oxalic acid and 0.5 mol/L oxalic acid,
123 respectively, and these values were consistent throughout the experiment. The Cs⁺ adsorbed on clay
124 minerals is considered to mainly take two forms (Fan et al. 2014). One is the outer-sphere complex on the
125 interlayer or planar site, and this form is relatively easy to desorb by alkali metal ions. The other cannot be
126 desorbed easily due to the formation of the inner-sphere complex on the interlayer or frayed edge site. The
127 Cs⁺, which was not desorbed by Li⁺, was considered to exist as an inner-sphere complex in biotite.

128 **Figure 1** shows the desorption efficiency of Cs^+ from biotite at each leaching time. LiNO_3 could desorb
129 approximately 31.6% of Cs^+ after 1 h of leaching time, and the desorption efficiency was consistent
130 throughout the experiment. According to a previous study, approximately 40% of Cs^+ in biotite is existed
131 as an ion-exchangeable form (Akemoto et al. 2021b). In this study, Li^+ desorbed the ion-exchangeable form
132 of Cs^+ from biotite and reached equilibrium within 1 h of the leaching time.

133 The desorption efficiency by oxalic acid increased with leaching time, and the value of 85.3% (0.1 mol/L)
134 and 94.9% (0.5 mol/L) were obtained after 2 weeks, and 89.8% (0.1 mol/L) and 96.1% (0.5 mol/L) were
135 obtained after 4 weeks of the experiment. These efficiencies were higher than that of Li^+ . The Cs^+ has been
136 considered to move into the interlayer of clay minerals and retain in an energy-stable site (Mukai et al.
137 2018). Since this study used biotite that has been saturated with Cs^+ and aged for 3 months, Cs^+ could have
138 occupied the energy-stable sites that were inaccessible. Therefore, the high efficiency of oxalic acid can be
139 attributed to the ability of oxalic acid to desorb Cs^+ in both outer-sphere complex and inner-sphere complex
140 with biotite.

141 Al, Mg, Fe, and Si are the main components of clay minerals, and the concentrations of each element in
142 the extract solution was measured as a function of leaching time. When LiNO_3 was used as an extraction
143 solution, Al, Mg, Fe, and Si was hardly observed in the solution (**Fig. S1**). **Figure 3a** indicates X-ray
144 diffractograms of biotite after each leaching time by LiNO_3 . The basal spacing of biotite after Cs^+ was 1.0
145 nm ($2\theta = 8.7^\circ$), indicating that Cs^+ was still adsorbed in the interlayer (Kogure et al. 2012). The intensity

146 of this reflection (8.7°) was consistent even after 4 weeks, the biotite structure was unaltered. This result
147 also indicated that LiNO_3 desorbed only Cs^+ in an ion-exchangeable form. In contrast, oxalic acid desorbed
148 Al, Mg, Fe, and Si in the biotite. The desorption rates of Al, Mg, and Fe were 68%, 83%, and 94% by 0.5
149 mol/L oxalic acid after 2 weeks, and this desorption rate did not significantly change until 4 weeks (**Fig. 2**).
150 Since the desorption efficiency of Cs^+ by 0.5 mol/L oxalic acid also plateaued at a leaching time of 2 weeks
151 (**Fig. 1**), these results indicate that the decomposition of clay minerals by oxalic acid (0.5 mol/L) induced
152 the desorption of Cs^+ from biotite at room temperature. When oxalic acid was 0.1 mol/L, the concentrations
153 of Al, Mg, and Fe continued to increase even after 4 weeks (**Fig. S1**). At 4 weeks of leaching time, the
154 desorption rates of Al, Mg, and Fe were 60%, 78%, and 91%, respectively.

155 Buckwheat has been reported to detoxify Al with oxalic acid released from its root, which forms an oxalate
156 complex, with Al to oxalic acid ratio of 1:3 (Ma et al. 1997). The reaction ratio between Fe and oxalic acid
157 was 2:5 (Lee et al. 2006). The concentration of Al and Fe in biotite used in this study was 3.2 mmol/g and
158 2.0 mmol/g, respectively. Therefore, even 0.1 mol/L of oxalic acid was sufficient to react with all Al and
159 Fe to form their complexes with oxalic acid. The XRD results indicated that the intensity of the biotite
160 reflection decreased with leaching time (**Figs. 3b and 3c**), although this biotite reflection (1.0 nm) has not
161 completely disappeared with 0.1 mol/L of oxalic acid, even at a leaching time of 4 weeks (**Fig. 3b**). This
162 result implied that the decomposition of biotite by 0.1 mol/L oxalic acid takes long time over 4 weeks. On
163 the other hand, it was almost completely disappeared with 0.5 mol/L oxalic acid after 2 weeks (**Fig. 3c**).

164 The broad peak around 11.5° was estimated to be oriented from a hydrotalcite-like structure, which has
165 XRD reflection at 11.7° and is similar to octahedral sheet (Miyata 1983; Bernard et al. 2022). Assuming
166 that Al and Fe eluted by oxalic acid leaching are metals that have existed in the tetrahedral sheet by
167 isomorphous substitute with Si, it is considered that the tetrahedral sheet could not maintain its structure
168 due to this elution. Since this reflection appears with the decrease in biotite reflection, it is presumed to be
169 from the remaining octahedral sheet. These results indicated that oxalic acid (0.5 mol/L) could desorb
170 elements (Al, Mg, Fe, and Si) in biotite and consequently decompose its structure. The Cs^+ desorption
171 from biotite due to decomposition of its structure by oxalic acid can be performed even without heating.

172 Oxalic acid is a natural organic acid present in various plants and is highly effective in the field of
173 environmental remediation (Huang et al. 2021; Costa et al. 2022). Since the results of this study confirmed
174 that Cs^+ strongly bound in clay minerals can be desorbed with oxalic acid without heating, there is the
175 possibility of introducing plants with oxalic acid content as a soil remediation technology option. Moreover,
176 this oxalic acid leaching has the potential to be a low energy soil remediation as it does not require heating.

177

178 **4 Conclusions**

179 In this study, the Cs^+ desorption from biotite using oxalic acid was investigated without heating. Oxalic
180 acid (0.5 mol/L) could desorb Cs^+ from biotite which has been aged for 3 months, and its desorption
181 efficiency was 94.9% after 2 weeks. In the same procedure, LiNO_3 could desorb only 32.2% of Cs^+ after 2

182 weeks. These results indicate the ability of oxalic acid to desorb Cs^+ from biotite not only the outer-sphere
183 complex like ion-exchangeable form but also the inner-sphere complex. Although the desorption of Cs^+
184 from biotite by oxalic acid leaching takes a long time (at least 2 weeks), the fact that no external energy is
185 required for extraction is a great advantage. Therefore, oxalic acid leaching without heating can be used for
186 the volume reduction of contaminated soil with radionuclides.

187

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193

194 **Author contribution**

195 **Yasuhiro Akemoto:** Conceptualization, Methodology, Formal analysis and investigation, Writing - original
196 draft preparation, Visualization

197 **Seira Takahashi:** Methodology, Investigation

198 **Toko Iwamura:** Methodology, Investigation

199 **Masahiko Kan:** Writing - review and editing

200 **Shunitz Tanaka:** Methodology, Writing - review and editing, Supervision

201

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204

205 **Availability of data and materials**

206 The datasets used and/or analyzed during the current study are available from the corresponding author on
207 reasonable request.

208

209

210 **Declarations**

211 **Competing Interests**

212 The authors declare that they have no known competing financial interest or personal relationship that could
213 have appeared to influence the work reported in this paper.

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274

275 **Figure captions**

276

277 **Fig. 1** Desorption efficiency of Cs⁺ from biotite at each leaching time at 20 ± 5 °C. Biotite: 0.05 g, the
278 leaching solution: 25 mL. Error bars indicate standard deviation (n = 3)

279

280 **Fig. 2** Desorption rate of elements at each leaching time from biotite by 0.5 mol/L oxalic acid; Symbols,
281 purple triangle was Fe, blue circle was Mg, red square was Al, and green rhombus was Si, respectively

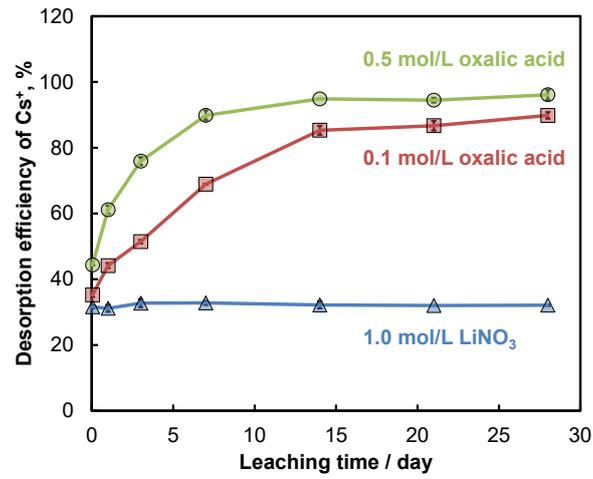
282

283 **Fig. 3** XRD patterns of biotite after each leaching treatment, a) 1.0 mol/L LiNO₃, b) 0.1 mol/L oxalic acid,
284 c) 0.5 mol/L oxalic acid

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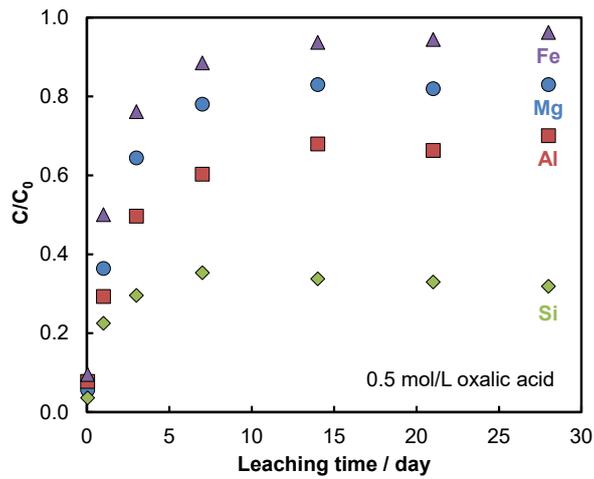


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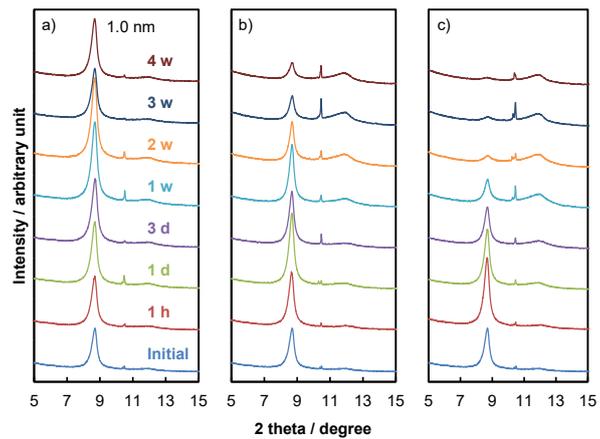


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300 c) 0.5 mol/L oxalic acid

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