Lysimeter tests for change in leachate quality from the modified removed soil: interaction between soil and polymer modification agent

Jialin Mo i), Kazuto Endo ii), Hiroyuki Arai iii), Takuya Miura i) and Kosuke Nakamura i)

i) Research Associate, National Institute for Environmental Studies, Fukushima Branch, Japan, 10-2 Fukasaku, Miharu, Tamura District, Fukushima 963-7700, Japan.
ii) Head, National Institute for Environmental Studies, Fukushima Branch, Japan, 10-2 Fukasaku, Miharu, Tamura District, Fukushima 963-7700, Japan.
iii) Specialist, National Institute for Environmental Studies, Fukushima Branch, Japan, 10-2 Fukasaku, Miharu, Tamura District, Fukushima 963-7700, Japan.

ABSTRACT

The removed soil generated from decontamination projects in Fukushima Prefecture is being transported to the interim storage facility (ISF) at present. In the ISF, modification agent such as water-absorbing polymer is used to improve handling of the removed soil during the separation process before storage. The expansion characteristic of the water-absorbing polymer agent might change the hydraulic property of the soil which will affect the leaching behavior. In addition, degradation of organic matter contained in the removed soil might influence the leachate quality as well. Thus, it is necessary to evaluate the environmental safety of the modified removed soil for safe storage in the ISF and its potential reuse in the future. In this study, two lysimeter tests on modified and unmodified removed soil was conducted to investigate its leachate quality. These tests lasted more than 400 days (liquid/solid ratios greater than 3.5). The highest $^{137}\text{Cs}$ concentration in leachate from the unmodified soil was 6.6 Bq/L at 250 days, and the accumulated leaching ratio was 0.09%. In leachate from the modified soil (addition of 3% water-absorbing polymer agent powder), the highest $^{137}\text{Cs}$ concentration was 3.3 Bq/L and the leaching ratio was 0.046%, a reduction by half. Leaching of $^{134}\text{Cs}$ from both soils was extremely low. The soil modification resulted in accumulated leaching of total organic carbon that was 60 % higher and leaching of $\text{K}^+$ and ammonia nitrogen (NH4-N) that was 35 % lower, respectively. No expansion of soil volume was observed after modification.

Keywords: removed soil, polymer modification agent, lysimeter test, leachate quality, radioactive cesium

1 INTRODUCTION

The nuclear accident happened at the Fukushima Daiichi Nuclear Power Plant on March 11, 2011 resulted in the contamination by radioactive substances such as cesium-134 and cesium-137 over a wide area, mainly in Fukushima Prefecture, Japan. (Ministry of the Environment of Japan (MOE) 2018).

Decontamination projects are being implemented in inhabitation areas of those affected regions since 2012. There are several methods including scraping topsoil for a few centimeters, removing sediments, branches and leaves, washing with water, etc. The removed soil and waste derived from decontamination projects were packed into container bags and stored in temporary storage sites at first. Since 2015, they are gradually being transported to the interim storage facility (ISF), which is located surrounding the Fukushima Daiichi Nuclear Power Plant. The total transportation volume is currently estimated at around 14 million m$^3$ and the transportation is expected to be completed by 2022 (MOE 2020).

The removed soil was transported to the Soil Separation Facilities in the ISF first, where it was taken out of the container bag and was separated from foreign materials over 2 cm such as plastic and plant. Then, separated soil was transported to the Soil Storage Facility where they were compacted and stored in the site with seepage control and leachate treatment equipment.

It was reported that the removed soil from farmlands contains large amount of cohesive soil mixed with organic matters which are hard to be separated (Inui et al. 2016). In addition, clogging of soil on the sieve will greatly reduce the separation efficiency in the ISF (Asada et al. 2018). Thus, modification agent such as water-absorbing polymer powder is applied to mix with the removed soil during separation process to improve the easiness of handling. Polymer agent can quickly absorb free water in the soil and become gelatinous after being added. The appearance of the modified removed soil will become loose and dry. As a result, it
is easier to separate foreign material such as weeds and roots and continuous handling of large volumes of the removed soil become possible.

Water-absorbing polymer has been widely used in hygienic and agricultural areas however its application in geotechnical filed is rare so far. There are a few studies focus on the soil structure after modification using X-ray CT scan (Tanaka et al. 2019, Shimizu et al. 2019). However, the physical and chemical properties of the modified removed soil have not been fully investigated and the impact of water-absorbing polymer agent remains unclear. The expansion characteristic of the polymer agent after absorbing water might change the hydraulic property of the soil which will affect the leaching behavior. In addition, degradation of organic matter in the removed soil might influence the leachate quality as well. Since the environmental safety is a crucial factor for the safe storage of the modified removed soil in the ISF and potential reuse of it as a geomaterial in the future, it is important to evaluate its leachate quality.

In this study, two lysimeter test were conducted on the original removed soil and the modified soil with addition of 3% water-absorbing polymer powder respectively. The water balance, displacement of the soil, leaching of radioactive cesium, leachate quality such as organic pollutant, nitrogen component and suspended solids was preliminarily discussed.

2 MATERIALS AND METHODS
2.1 Removed soil and polymer modification agent

The removed soil was obtained from a temporary storage site in Inata village, Fukushima Prefecture. The radioactivity level of $^{134}$Cs and $^{137}$Cs, which is the major contaminants, was measured before the test (Jan.14, 2019) by gamma-ray spectroscopy with germanium (Ge) semiconductor detector.

Polymer modification agent used in study was supplied from Leafair Co., Ltd. It is a kind of water-absorbing polymer powder and the initial water content is around 10%. Its swelling property was estimated based on the Japan Bentonite Manufacture Association Standard (JBAS-104-77). 2 g agent was added into a 100 mL measuring cylinder filled with 100 mL distilled water by 10 times. Entire 100 mL distilled water became gelatinous immediately after addition. Thus, another case was conducted by adding 0.06 g agent into 100 mL distilled water. The volume of the generated gelatinous material was 12 mL after sitting undisturbed for 24 hours, suggesting that 1 g agent can absorb 400 mL water and turn it into gel.

The water-absorbing polymer powder was added into the removed soil by 3% of the natural weight of the soil for the modification. The removed soil and polymer agent were mixed using a mortar mixer after weighing. The properties of the removed soil and the soil-agent mixture were shown in Table 1.

| Table 1. Properties of the removed soil and soil-agent mixture. |
|-----------------|-----------------|-----------------|
| Items | Standard/Method | Removed soil | Soil-agent mixture |
|-----------------|-----------------|-----------------|
| Radioactivity level | $^{134}$Cs | 11704 Bq/kg | 10794 Bq/kg |
| | $^{137}$Cs | 936 Bq/kg | 868 Bq/kg |
| | spectrometry using Ge detector | 10768 Bq/kg | 9926 Bq/kg |
| Particle density | JIS A 1202 | 2.33 g/cm$^3$ | 2.49 g/cm$^3$ |
| Water content | JIS A 1203 | 48.6% | 46.9% |
| C, H, N, S content | vario MACRO cube | 6.49% | 6.36% |
| | | 1.20% | 1.17% |
| | | 0.42% | 0.39% |
| | | 0.06% | 0.07% |
| Ignition loss | JIS A 1226 | 15.78% | 15.62% |
| Total organic carbon content | JIS K 0102 | 95.3 g/kg | 100.4 g/kg |
| Maximum dry density | JIS A 1210 | 1.09 g/cm$^3$ | 1.02 g/cm$^3$ |
| Optimum water content | | 39.5% | 27.3% |
| Hydraulic conductivity | JIS A 1218 | $3.15 \times 10^{4}$ cm/s | $1.06 \times 10^{4}$ cm/s |

2.2 Lysimeter test

The two lysimeters (2 m × 2 m × 2 m each) were set up in the laboratory where the ambient temperature is around 20°C. The leachate was collected in a tank connecting the bottom center of the lysimeter through a polyvinyl chloride pipe. Glass beads and stainless-steel mesh was set over the pipe to prevent leaching of soil particles. A rainfall simulator was set over the top of the lysimeter with 400 needles evenly distributed to provide water.

The removed soil was compacted in the lysimeter in 5 layers and the modified soil was compacted in 4 layers. The compaction degree was set to 75% of the maximum dry density and the total height of the soil was set to 1.16 m. The wet density and the porosity of the removed soil after compaction was 1.203 Mg/m$^3$ and 0.65 respectively. For the modified soil, the wet density was 1.147 Mg/m$^3$ and the porosity was 0.64.

Fig. 1. Schematic view of lysimeter test.
For both lysimeters, a 0.5-meter thick layer of clean decomposed granite soil was compacted under the compaction degree of 80% on the top to reduce the air radiation level. The schematic view of the lysimeter test was shown in Fig. 1.

The two lysimeters were left standing for 25 days after compaction and then the rainfall simulation started. The rainfall was set to 8 mm/day (32 L) and the flow rate was 1.6 L/min for 20 minutes each day. On every Tuesday morning, all the leachate stored in the tank under each lysimeter was pumped up into the tank on the ground and sampled for analysis of leaching of $^{134}$Cs and $^{137}$Cs, leachate quality such as pH, EC, Eh, TOC, DOC, BOD, COD, SS, etc.

3 RESULTS AND DISCUSSION

3.1 Settlement and water balance

Settlement of soil and accumulated leachate amount of both lysimeter tests are shown in Fig. 1.

The distance between the top of the soil and the ground surface was measured at four sides and center of the lysimeter. The ground level was set as 0 m and the average level of the five measuring points was used as the surface height of the soil. Settlement of the removed soil started right after rainfall simulation began. However, no obvious settlement was observed for the modified soil in the first 28 days after rainfall simulation started. It was probably because that the polymer agent absorbed water and swelled, which mitigated the settlement at the initial stage. The settlement of both soils became stable after 300 days and the surface height was similar. Modification of the removed soil with polymer agent has no obvious influence on the soil volume change in the long term.

The accumulated rainfall for both lysimeters was 3360 mm (around 13440 L). For the removed soil, 70 L of the leachate was firstly collected after 21 days of rainfall. For the modified soil, it took 28 days to firstly collect 55 L of the leachate. The removed soil retained 600 L of water to reach field capacity and 40% additional water was retained after modification. 200 L of the leachate can be collected from the removed soil every week and the surplus of 24 L of rainfall might be evaporated. For the modified soil, the total volume of leachate collected after 400 days was around 600 L less compared to the removed soil. Thus, the polymer agent in the soil might retain additional water.

3.2 Leachate quality

The variation of pH, electrical conductivity (EC) and redox potential (Eh) of the leachate with time of both lysimeter tests are shown in Fig. 3, Fig. 4 and Fig. 5, respectively. The initial pH of both soils was between 6.5 to 7.0 and gradually increased to 7.0 to 7.5. There was a sudden decrease of pH after 350 days and the reason was not clear.

The EC of the leachate from the modified soil was

![Fig. 2. Settlement of soil and accumulated leachate amount.](image)

![Fig. 3. Variation of pH with time.](image)

![Fig. 4. Variation of electrical conductivity (EC) with time.](image)

![Fig. 5. Variation of redox potential (Eh) with time.](image)
higher than the removed soil case before 300 days, which was probably caused by the leaching of Na\(^+\) and SO\(_4^{2-}\) from the polymer agent. As shown in Fig. 6 and Fig. 7, the leaching of Na\(^+\) and SO\(_4^{2-}\) from the removed soil was less than 20 mg/L without obvious change. However, the highest concentration of Na\(^+\) and SO\(_4^{2-}\) leached from the modified soil was over 500 mg/L and 700 mg/L at around 150 days. The leaching of Na\(^+\) gradually decreased after 200 days and its variation trend was similar with EC.

The variation trend of the Eh for both soils was similar. The Eh started to decrease at 100 days from 100 mV and became stable around -50 to -100 mV after 200 days. The pore volumes of flow (PVF) reached to 1 during this stage and the microbial activity might be higher along with the distribution of water in the soil, which leaded to the decrease of the Eh.

The variation of total organic carbon (TOC) and dissolved organic carbon (DOC) with time are shown in Fig. 8. There was no obvious difference between the TOC and DOC, suggesting that most of the organic carbon content dissolved in leachate. For the removed soil, leaching of TOC gradually increased and became stable around 100 mg/L after 200 days. For the modified soil, leaching of TOC was higher and increased before 200 days as well. However, concentration of TOC kept decreased after 200 days and became lower than the removed soils case after 300 days. The accumulated leaching of TOC from the modified soil was 1.6 times higher and additional organic carbon might be leached out from the polymer agent. In addition, polymer agent absorbs free water and introduced the aggregation of fine particles which might reduce the leaching of organic carbon from the soil itself.

The variation of total nitrogen (T-N) and dissolved total nitrogen (DT-N) with time are shown in Fig. 9. Most of the total nitrogen content was soluble which was similar with the organic carbon. Concentration of T-N of both cases kept decreasing before 120 days when the PVF reached to 1 and the main content was nitrate nitrogen (NH\(_3\)-N) based on the analysis results. Thus, soluble NH\(_3\)-N existed in the pore spaces was washed out during this stage. Concentration of T-N after 120 days gradually increased and then kept stable around 20 mg/L. The main content during this stage was ammonia nitrogen (NH\(_4\)-N) as shown in Fig. 16.

The variation of chemical oxygen demand (COD) and biochemical oxygen demand (BOD) with time are shown in Fig. 10 and Fig. 11. The variation trend of COD was similar with the results of the TOC. The concentration of BOD in the leachate from the removed soil gradually increased from 120 days and reached to 50 mg/L at 190 days. The soluble content of BOD in the pore space might be washed out during this stage. For the modified soil, the concentration of BOD suddenly increased from 10 mg/L to 200 mg/L at
around 200 days and then decreased quickly in the next 50 days. Addition of polymer agent might mitigate the time for soluble BOD content to be washed out from the soil. However, the reason for the high concentration of BOD shown in the leachate at 200 days was not clear.

The variation of suspended solid (SS) with time is shown in Fig. 12. For the modified soil, almost no SS leached out until 300 days. It was probably because the gelatinous substance generated by polymer agent after absorbing water aggregated fine particles and mitigated the leaching of SS from the soil. The leaching of SS from the removed soil started increasing after 200 days and the concentration of SS in the leachate of the modified soil also increased after 300 days. Considering that there was weeds and roots existing in the removed soil, the leaching of organic matter as SS might happen.

3.3 Leaching of radioactive cesium

The radioactivity level of $^{134}$Cs and $^{137}$Cs of the leachate was both measured by the γ-ray spectroscopy with Ge semiconductor detector. Since the radioactivity level of $^{134}$Cs was extremely low, only the results of $^{137}$Cs are shown in Fig. 13.

For the removed soil, the leaching of $^{137}$Cs kept increasing to 6.6 Bq/L at 250 days and then gradually decreased. For the modified soil, leaching of $^{137}$Cs kept increasing in the first 150 days which was similar with the removed soil. Then the leaching of $^{137}$Cs started decreasing from 3.3 Bq/L to 1.8 Bq/L until 300 days when the concentration increased to 2.8 Bq/L at 340 days and then decreased again. The highest leaching concentration of $^{137}$Cs from the removed soil decreased by 50% after being modified with 3% polymer agent. In the first 150 days, the leaching of $^{137}$Cs was similar between two soils. Considering that the PVF reached to 1 at this stage, the polymer agent distributed in the soil might have not fully contacted with the infiltrated water yet thus its impact on the leaching was not obvious. For the modified soil, the increased leaching of $^{137}$Cs after 300 days was at the same time when leaching of SS increased. The additional leaching of $^{137}$Cs might be the $^{137}$Cs absorbed on the organic matter which leached out as SS at this stage.

The leaching ratio of $^{137}$Cs of both soils are shown in Fig. 14. The leaching ratio of $^{137}$Cs from the removed soil and the modified soil was 0.09% and 0.046% and kept increasing. The accumulated leaching amount of $^{137}$Cs from the modified soil was half of the leaching amount from the removed soil and the increasing of leaching ratio became obviously lower after 200 days. Ishimori et al. (2017) suggested that the existing of potassium ion ($K^+$) and ammonium ion ($NH_4^+$) significantly affects the desorption of radioactive Cs. The variation of $K^+$ and ammonia nitrogen ($NH_2-N$) with time are shown in Fig. 15 and Fig. 16. The concentration of $K^+$ and $NH_2-N$ in the leachate from the
removed soil was higher than the modified soil case. In addition, the accumulated leaching amount of K\(^+\) and NH\(_2\)-N from the removed soil was 1.4 and 1.6 times higher, respectively. The higher concentration of both ions in the leachate might stimulate the leaching of \(^{137}\)Cs from the removed soil. The polymer agent and the generated gelatinous substance might also have certain adsorption ability on \(^{137}\)Cs thus its concentration in the leachate from the modified soil was lower.

4 CONCLUSIONS

This research was conducted to evaluate the impact of polymer modification agent on the leachate quality from the removed soil in Fukushima. Lysimeter tests using the original removed soil and modified removed soil mixing 3% water-absorbing polymer agent powder were conducted.

The highest \(^{137}\)Cs concentration in leachate from the original removed soil was 6.6 Bq/L at 250 days and the accumulated leaching ratio was 0.09%. Addition of 3% water-absorbing polymer agent powder into the soil reduced the highest leaching concentration of \(^{137}\)Cs to 3.3 Bq/L and the leaching ratio to 0.046%, a reduction by half.

The water balance and the settlement of the soil in two lysimeters were similar. No expansion of soil volume was observed even though additional water was absorbed by polymer agent in the modified soil.

The accumulated leaching of total organic carbon from the modified soil was 60% higher and the leaching of K\(^+\) and NH\(_2\)-N was 35% lower. The higher K\(^+\) and NH\(_2\)-N concentration in the leachate from the removed soil without modification might stimulate the leaching of \(^{137}\)Cs. In addition, adsorption of \(^{137}\)Cs on the water-absorbing polymer powder might also mitigate its leaching and further studies is necessary to evaluate the leaching mechanism of \(^{137}\)Cs affected by the polymer agent.

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