DEVELOPMENT OF MUD-CRUSHING METHODS FOR FARMLAND-DERIVED CLAYEY SOIL AND THEIR APPLICATION TO PARTICLE-SIZE SEPARATION FOR DECONTAMINATION OF CESIUM-CONTAMINATED SOIL

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The volume of radioactive cesium-contaminated soil being removed during the cleanup process after the explosion of Fukushima nuclear power plant 1 is estimated to be 13 million m³. Soil contaminated with more than 20,000 Bq/kg of radioactivity is targeted for decontamination by the particle-size separation method to pass the criteria for reuse in Japan. Because more than 80% of the targeted soil is composed of fine silt and clay derived from farmland, a system was developed to increase the amount of recyclable soil by a combination of mud crushing and fine-particle separation with the classification point set lower than about 20 μm. Mud crushing is one of the key technologies for effective decontamination because clay forms aggregates due to the presence of organics such as humus. Four types of mud-crushing equipment were examined on site, followed by pebble milling or ultrasonic dispersion, both of which showed promising results. Using the selected equipment, the proposed soil-size separation was carried out. Overall, the proposed system was proven effective by the results of radioactivity reduction ratio and volume of soil for recycling. Furthermore, lab-scale pebble mill tests using a pot-type wet mill were carried out to confirm the basic characteristics of mud crushing for organic aggregates.

Key Words: cesium-contaminated soil, mud crushing, particle size separation, pebble milling, ultrasonic disperser

1. INTRODUCTION

According to the Japan Ministry of the Environment (MOE), an estimated 13 million m³ of soil contaminated with radioactive cesium 134 and 137 was removed during the process to clean up the damage from the accident at Fukushima nuclear power plant 1. The Ministry plans to store this soil at interim storage facilities at Okuma and Futaba, Fukushima Prefecture. Because they are mandated to be removed until 2045, it is highly important to minimize the volume of soil that is taken to the final disposal site by applying some method of decontamination.

Wet type soil particle-size separation, commonly used for purification of soil contaminated with heavy metals, is considered to be a promising method. Using the fact that most of the radioactive cesium is strongly retained by the clay minerals in the fine-grained fraction and does not easily desorb, separation of the cesium-contaminated clay/silt particles reduces the radioactivity concentration of the sand/gravel fraction, making it suitable for reuse as soil material. Typically, the classification point is 75 μm.

However, the soil being removed during remediation that will require subsequent volume reduction has a radioactivity concentration of 20,000 Bq/kg to 80,000 Bq/kg (these values are as of March, 2015).
More than 80% of this soil is farmland-derived clayey soil containing fine particles smaller than 75 μm, and most of this soil remains in the contaminated fraction after normal particle-size separation treatment with a classification point of 75 μm\(^1\). This fact leads to the fundamental problem of difficulty of achieving sufficient volume reduction. To solve this problem, the authors propose a particle-size separation system that reduces the classification point down to 20 μm, which reflects the minimum capacity of hydro-cyclones in the field of civil engineering. Also, from a phenomenal point of view, soil particles of 20 μm or less are likely to have the problem of reaggregation according to the soil particle-size classification by the International Society of Soil Science\(^1\). We call this method the “fine particle-size separation system” (FPSSS). The FPSSS approach can be expected to produce a processing system that extends general particle-size separation processing to increase the recycling rate without adding significant additional cost.

However, particle-size separation itself presents problems when applied to farmland-derived clayey soil, such as aggregation by organic matter, ineffective consumption of chemicals used during processing, contamination of organic matter in the concentrate after separation, inability to dehydrate the fine soil particles after classification. The effect of modifiers applied for sorting, was already explored\(^10\). In each case, sufficient research has not been carried out yet, and solving these problems would lead to a breakthrough in decontamination and reuse technology for radioactive cesium-contaminated soil. Among these, aggregation by organic matter was the focus of this study.

Farmland-derived clayey soil contains organic matter, such as aggregation by large soil particles such as sand and gravel as shown in Fig. 2. Volume decomposition decomposes the humus-aggregated particles as shown in Fig. 2(b). These modes are considered to differ depending on the properties of the object to be decomposed, the properties of the medium used, and the force applied. To make the proposed FPSSS effi-

![Fig. 1](image1.png) Schematic drawings of clay aggregate formed by the presence of organic matter.

![Fig. 2](image2.png) Schematic drawings of decomposition modes for soil aggregates: (a) Surface decomposition, (b) Volume decomposition.

### 2. PURPOSE OF THIS STUDY

The purpose of this study is first, to develop a method for decomposing organic aggregates that adhere to gravel/sand particles, which is necessary to increase the decontamination rate and increase the recycling rate/amount by FPSSS. Then, the extent to which the decontamination rate and recycling rate/amount can be improved by applying a combination of the selected decomposition method and FPSSS was evaluated.

Therefore, four types of mud-crushing (MC) equipment were selected and tested on site. Their application conditions were optimized using actual soil, and a comparative study was conducted under these conditions to select appropriate equipment for subsequent evaluations. Second, from the viewpoint of reducing the radioactivity concentration, the effect of the treatment system by the combination of MC with selected equipment and FPSSS of actual contaminated soil was confirmed. After this series of experiments, a study using a pot-type wet mill was carried out to confirm the basic properties of decomposition on farmland soil that was aggregated by the action of humus.

Here, it is considered that there are two modes of decomposition, as shown in **Fig. 2**. Surface decomposition removes fine particles adhering on the surface of relatively large soil particles such as sand and gravel as shown in **Fig. 2(a)**. Volume decomposition decomposes the humus-aggregated particles as shown in **Fig. 2(b)**\(^16\). These modes are considered to differ depending on the properties of the object to be decomposed, the properties of the medium used, and the force applied. To make the proposed FPSSS effi-
cient, a MC method is required to transfer clay minerals containing radioactive cesium through a sieve (20 μm or less) without destroying the sand and gravel particles that we want to retain for recycling. This requires both surface crushing of the organic aggregates adhering to the surface of sand and gravel and volume crushing of the organic aggregates.

3. MATERIALS AND METHODS

(1) Materials
As shown in Table 1, the soil samples used in the tests were collected from farmland in Ibaraki Prefecture near Fukushima Prefecture and from Fukushima Prefecture. Note that Soil AF was not directly collected from fields but was provided by the intermediate storage facility, and its source is unspecified. Table 2 shows the particle-size distribution of each sample according to JIS A 1204. Table 3 lists the results of analyzing four properties: ignition loss (JIS A 1226), total organic content (TOC, JIS K 0102 22), humic acid content, and fulvic acid content. The humic and fulvic acid content were analyzed by NaOH fractionation and the dichromic acid colorimetric method to determine the organic matter content (JSF E 232-1988). Commercially available Andisol (Soil A) was also used for the lab test as a sample of highly aggregated soil. The particle-size distribution for Soil A was analyzed by wet sieving for particles larger than 250 μm and by a laser diffraction particle-size distribution analyzer (Shimadzu, SALD-2300) for particles smaller than 250 μm.

The uncontaminated soil collected from Ibaraki contained a large amount of fine particles and a high-organic matter content compared to contaminated soil from Fukushima. However, the content of humus, such as humic acid and fulvic acid, did not differ as much as the total organic matter content. Table 4 shows the dry base radioactive cesium concentration of each contaminated soil measured by a Ge semiconductor detector (ORTEC, GEM 20P4-70). In addition to the radioactive cesium concentration of the original soil, the radioactive concentration of the slurry after classification at 75 μm was also measured for the solids after centrifuging.

(2) Selection of mud-crushing equipment
From the results of literature surveys, preliminary tests, and the feasibility of procurement for actual projects, four types of MC equipment were selected: pebble mill, ultrasonic disperser, high-pressure ejector, and super shear mixer. The characteristics of each are described below.

a) Pebble mill
Pebble milling decomposes aggregates using pebbles as a medium within a rotating body that causes the pebbles and aggregates to collide in a muddy water. According to the basic experiments described in Section 4(4), pebbles (gravels) of several millimeters

| Symbol | Category | Sampling place | Field category |
|--------|----------|----------------|---------------|
| X      | Farmland soil | Omitama, Ibaraki | Sweet potato farm |
| Y      | Farmland soil | Daigo, Ibaraki | Rice paddy |
| Z      | Farmland soil | Hitachinaka, Ibaraki | Farmland |
| N      | Farmland soil | Okuma, Fukushima | Rice paddy |
| S      | Farmland soil | Okuma, Fukushima | Rice paddy |
| AF     | Unspecified removed soil | Unspecified | Unspecified |
| A      | Andisol | Kanuma, Tochigi | Unspecified |

| Property | Uncontaminated | Contaminated | Ref. |
|-----------|----------------|--------------|------|
| Particle density (g/cm³) | 2.64 | 2.40 | 2.66 |
| Gravel, 2-75 mm (%) | 0.1 | 0.4 | 0.1 | 11.5 | 4.7 | 18.4 | 1.5 |
| Sand, 0.057-2 mm (%) | 21.9 | 22.3 | 25.0 | 46.1 | 21.3 | 45.9 | 96.7 |
| Silt, 0.005-0.075 mm (%) | 70.9 | 63.2 | 65.7 | 32.5 | 42.0 | 27.8 | 1.7 |
| Clay, <0.005 mm (%) | 7.1 | 9.9 | 9.2 | 9.9 | 32.0 | 7.9 | 0.1 |
| Maximum particle size (mm) | 4.8 | 9.5 | 4.8 | 19.0 | 19.0 | 26.5 | - |
| Uniformity coefficient Uc | 8.1 | 8.8 | 10.2 | 45.8 | 51.3 | - |

| Characteristic | X | Y | Z | N | S | AF |
|----------------|---|---|---|---|---|----|
| Vol. water content (%) | 34.7 | 44.9 | 35.6 | 23.8 | 34.9 | 23.2 |
| Ignition loss (% dry) | 18 | 25 | 18 | 8.7 | 13 | 8.37 |
| TOC (% dry) | 2.78 | 7.67 | 2.51 | 1.9 | 3.3 | 2.46 |
| Humic acid (% dry) | 0.69 | 4.43 | 0.58 | 1.20 | 4.08 | 0.90 |
| Fulvic acid (% dry) | 0.97 | 2.04 | 0.89 | 0.66 | 0.85 | 0.58 |
| Slurry pH (-) | 9.8 | 9.6 | 9.3 | 6.5 | 6.1 | 7.1 |

| Material | N | S | AF |
|-----------|---|---|----|
| Cs-134 (Bq/kg dry) | 394 | 184 | 195 |
| Cs-137 (Bq/kg dry) | 6,168 | 2,611 | 2,863 |
| Total (Bq/kg dry) | 6,562 | 2,795 | 3,058 |
| Slurry < 75 μm (Bq/kg dry) | 12,400 | 3,600 | 8,059 |

All data measured in 2017.
are effective as a medium for soft organic aggregates. In this experiment, a drum washer with a rotating body 586 mm in diameter and 935 mm-long (Kikosha, batch-type), with a volume of about 45 L, was used (Fig. 3). Pebble media taken from mountainous areas with a grain size adjusted to 4 mm was used.

The optimum rotation speed is given in the manufacturer’s specifications, which is 70 % of critical rotation speed for the machine (Kikosha, KDW2142) supposed to be used in the real project\(^{18,19}\). Using the formula to calculate the centrifugal acceleration, \[1.118 \cdot r \cdot n^2 \cdot 10^{-6} \cdot G\] (\(r\):radius, \(n\):rotation speed, \(G\):gavity), it yields as 0.47 G and the rotation speed for the small-type mill used in this experiment is calculated accordingly as 37.9 rpm.

The standard exposure time was set by assuming that the drum-moving distance was the same as that of a production machine (diameter 2.1 m and processing capacity of 150 t/hr at 20 rpm), which is 2 minutes if the number of shearing collisions is considered to be almost the same and the moving distance is the same as the entanglement. In this way, 1 unit of standard exposure time (indicated as 1 repetition, or rep, hereafter) was determined as 3 min and 47 s (37.9 rpm, diameter 586 mm) for this test machine.

b) Ultrasonic disperser

An ultrasonic disperser causes surface destruction by strong cavitation by high-power ultrasonic waves. Because the ultrasonic efficiency is 96%, heat generation is low. High output up to 16,000 W is possible. This study used a machine made by Heelsher Ultrasound GmbH with an output of 2.0 kW, frequency of 20 kHz, and processing capacity of 200-300 L/hr (Fig. 4).

c) High-pressure water ejector

As illustrated in Fig. 5, a high-pressure water ejector applies high-pressure air and liquid simultaneously to a slurry in the primary ejector section to promote destruction by cavitation. The secondary ejector section decomposes the remaining aggregates at the collision plate in the end. The primary ejector diameter is 50 mm, discharge pressure is 14 MPa, processing capacity is 7 L/min (when pressure is 4.5 MPa), and power is 30 kW. The discharge pressure can vary from 4 MPa to 12 MPa, and the maximum pressure of 12 MPa was used in this study. Shoda et al. have confirmed its effect for the bottom mud of reservoirs\(^{20,21}\).

d) Super shear mixer

A super shear mixer is composed of a mixing head unit and a power unit, and the mixing head unit is composed of a rotor and a stator. As shown in Fig. 6 (right), the rotor is rotated at a high speed to pull the slurry through 1/4 spherical indentations, where a shearing force is exerted. Further, the shearing by the high-speed vortex formed on both sides of the gap promotes crushing. The equipment used was a 060M1.5 mixer manufactured by Satake Chemical Equipment Mfg., Ltd., with a processing capacity of 1.9 m\(^3\)/hr and a power of 1.5 kW. The frequency can be changed from 20 Hz to 60 Hz, and the most effective frequency of 60 Hz was used.

\(\text{(3) Methods}\)

Experiments were conducted on site in three steps: (a) optimization of operating conditions for MC equipment, (b) validation of four types of MC equip-
ment under the optimum conditions, and (c) evaluation of the effects of reducing the radioactivity concentration using actual contaminated soil.

After this series of site-based experiments, additional lab-scale tests were conducted to examine the organic matter migration and decomposition mode using a pot-type pebble mill.

a) Optimization of operating conditions for mud-crushing equipment

Mud-crushing tests were conducted on the four selected types of equipment to confirm the optimum operating time, the number of repetitions, and applicability to soil derived from farmlands. Three types of uncontaminated soil (X, Y, and Z) and actual contaminated soil (AF) were used as shown in Table 1, each of which was passed through a 2-mm wet vibrating screen and then tested with a slurry of 75 μm or less by elutriation. Calcium hydroxide [Ca(OH)₂] was added to the slurry in accordance with previous research to obtain a pH of around 10.5 to prevent the elution of humus and reaggregation of negatively charged soil particles. The particle-size distribution was measured using a laser diffraction particle-size distribution analyzer (Shimadzu Corp., SALD-2300).

b) Validation of mud-crushing equipment

Under the conditions defined in the previous section, MC tests were conducted using three types of uncontaminated soil and three types of contaminated soil. The pH of the slurry to be tested was adjusted to about 10.5 in the same manner as in a). The MC tests were performed twice each and averaged. Due to equipment use restrictions for the high-pressure ejector and super shear mixer, some tests could not be performed using actual contaminated soil.

c) Evaluation of the effects using contaminated soil

To confirm the transition processes of soil particle size and radioactivity concentration, a series of experiments was carried out using three types of actual contaminated soil with the MC equipment and operating conditions selected by the above tests. Soil particles in the produced slurry were classified at 20 μm or less using a hydro-cyclone separator (Kikosha, TC-3). The total amount of each slurry fraction was measured after the slurry was centrifugally separated. The radioactivity concentration was measured with a Ge semiconductor detector (ORTEC, GEM 20P4-70).

d) Examination of organic matter migration and decomposition mode in the pebble mill

For the pebble mill and ultrasonic disperser, whose effectiveness was confirmed by the tests in the previous section, the basic properties of the soil after decomposition were confirmed for the pebble mill, which has advantages in terms of procurability and applicability. It is assumed that the aggregates due to the intervention of organic matter with low adhesive force will be decomposed first. Tests were conducted to confirm if changes in the distribution of particle size and migration of organic matter to estimate selective decomposition of organic aggregates that loosen only the bonds due to humus without destroying the particles themselves, such as sand, can be achieved.

Commercially available Andisol (Kanuma Andisol) was prepared as a sample containing relatively more organics that form aggregates. A porcelain pot with a diameter of 130 mm and an internal volume of 900 mL was used (Fig. 7), and 107 g of Andisol (average water content 31%), 500 mL of water, and 300 mL of crushing medium were added thereto. Alumina balls with a diameter of 6 mm or 10 mm were used as the decomposition medium. The rotation speed was 24 rpm (pot peripheral speed of 16.3 cm/s), and the mill was operated at room temperature. After MC, sieving was performed by wet vibration sieving, and then the weight and organic matter content were measured for each fraction. Because ignition loss is considered to include inorganic carbon content and absorbed water, an energy dispersive X-ray fluorescence analyzer (XRF, Shimadzu Corp., EDX-8000) was used as a simple method to measure organic matter content. The inorganic elements of the sample were identified in the qualitative mode, and the elements with an atomic number of 24 or more (Mg or higher) were detected by qualitative quantitative analysis with a high content of about 98% or more of the inorganic substances. Organic matter (C₆H₁₀O₅·4H₂O) was added to balance this, and in addition to the measurement of the abovementioned elements with higher atomic number (of the predetermined analysis line) using the quantitative mode, all indefinite analysis lines were displayed as balance percent derived from organic matter, thus contain organic matter. The amount percentage was estimated and converted to TOC percent. Because it was confirmed that the estimated TOC value obtained in this way was almost the same as the value directly measured by a TOC meter (with solid sample combustion device), this method was used.

Fig. 7 Porcelain pot mill and alumina ball.
In addition, a porcelain pot with a diameter of 215 mm and an internal volume of 4800 mL, Asake sand (specific density 2.6 g/cm³) with a diameter of 5 mm as a crushing medium, and paddy soil Y (see Table 1) were prepared. The reason for using Asake sand here is that sand can be easily procured anywhere. Before milling, 500 mL of water and 300 mL of 5 mm Asake sand were added to 107 g of soil, and then MC was conducted at a rotation speed of 45 rpm (peripheral pod speed of 50.6 cm/s). In Runs 5 and 6, pH was set to 11.5 using Ca(OH)₂ to confirm the effect as described above. Furthermore, in Run 6, to confirm the effect of immersion before MC, the target soil, water, Asake sand, and Ca(OH)₂ were mixed, and this was allowed to stand in a constant-temperature bath at 45°C for 10 minutes before MC by the pebble mill.

4. RESULTS AND DISCUSSION

(1) Optimization of operating conditions for mud-crushing equipment

To confirm the optimum operating time and number of repetitions for each type of decomposition equipment, a test was conducted from the viewpoint of how much the target soil particles ≤20 µm increase. As shown in Fig. 8, for the pebble mill and ultrasonic disperser, the larger the number of repetitions, the higher the decomposition effect. It was determined that 5 reps (= 18 min 55 s) in the pebble mill and 15 reps (= 22.5 s) in the ultrasonic disperser were optimal. Since the rate of increase in Fig. 8 becomes almost stable and small between 12 reps and 18 reps while restricting energy consumption as low as possible, 15 reps in the ultrasonic disperser is selected. Also, because there was no significant difference between the high-pressure ejector and the shear mixer as the number of repetitions increased, 2 reps and 3 reps were determined to be optimal, respectively, considering the change in particle-size distribution.

(2) Validation of mud-crushing equipment

Changes in particle-size distribution are shown in Figs. 9 and 10, and changes in the amount of soil particles in the fractions of 20-75 µm and <20 µm are shown in Table 5. The shear mixer could not be tested on actual contaminated soil. In these figures, thin curves show the particle-size frequency before MC and thick curves show the frequency after MC.

From Fig. 9, Fig. 10, and Table 5, it can be seen that the soil particles of 20-75 µm were decomposed and reduced to 20 µm or less for all types of soils except Soil AF for both the pebble mill and ultrasonic disperser. However, the MC effect was hardly seen for the high-pressure ejector and the super shear
Fig. 9 Soil particle-size distribution before and after MC with each machine for Soils X (left), Y (center), and Z (right). Pebble mill (a), Ultrasonic disperser (b), High-pressure water ejector (c), Super shear mixer (d). Thin curves are for before MC and thick curves are for after MC.

Fig. 10 Soil particle-size distribution before and after MC with each machine for Soils N (left), S (center), and AF (right). Pebble mill (a), Ultrasonic disperser (b). Thin curves are for before MC and thick curves are for after MC.
Because the ultrasonic disperser is thought to be superior in pulverizing organic matter, it was expected that the decomposition products from the ultrasonic disperser would contain finer soil particles. This assumption was confirmed by the experimental results, which showed particle sizes less than 1 μm, were more remarkable for the ultrasonic disperser than for the pebble mill.

Regarding Soil AF, almost no decomposition effect was observed for all four MC types. From these results, Soil AF can be considered as not derived from farmland even if Soil AF contained a certain amount of organic matter and showed a lower silt and clay content.

(3) Evaluation of the effects using contaminated soil

Tables 6-8 show the dry weight, radioactivity concentration, and radioactivity of the particles measuring 20-75 μm and <20 μm after MC was performed on Soils N, S, and AF. Values for >75 μm are not directly measured, but calculated from the weight fraction and radioactivity concentration of the original soil. In Soil N, the dry weight fractions of soil particles of 20-75 μm decreased from 10% to 4% and 7% respectively, and the radioactivity fraction also decreased from 15% to 3% and 7% for the pebble mill and ultrasonic disperser, respectively. In Soil S, the dry weight fractions of soil particles decreased...
from 15% to 6% and 8% for the pebble mill and ultrasonic disperser, respectively. The corresponding radioactivity fraction decrease was from 13% to 5% and 5%, which means that the volume of soil particles <20 µm increased due to the progressive decomposition of aggregates containing clay particles. This shows that radioactive cesium was successfully concentrated in the finest fraction by both the pebble mill and the ultrasonic disperser. It is thought that the 2:1 clay adsorbing radioactive cesium steadily shifts to the fine particle fraction due to the combined occurrence of volumetric and surface decomposition, as illustrated in Fig. 2.

In contrast, regarding Soil AF, Table 8 shows almost no change in both the particle-size distribution and in the radioactivity fraction before and after MC. As discussed above, it can be concluded that this soil does not form organic aggregates, which are the target of MC.

(4) Examination of organic matter migration and decomposition mode using pebble mill

Table 9 shows the test conditions for Runs 1-6. Figure 11 shows the change in the soil particle-size distribution before and after MC by a porcelain pot mill. The distribution peaked in the 106- to 710-µm fraction (black in the figure) without MC and this peak moved to a finer grain size (in the order of red, green, and blue from original soil) after MC due to decomposition. When compared in terms of MC time, the distribution shifts more to the left side after 40 minutes than after 10 minutes with 10-mm balls (red and green in the figure). When compared in terms of ball size, the distribution with 6-mm balls shifted to the left side slightly more than it did with 10-mm balls (blue in the figure).

It should be noted that both the volumetric decomposition (compression crushing by impact or compression) and surface decomposition (shear crushing, such as by friction) shown in Fig. 2 occur at the same time due to the fact that the distribution peak moves to the finer side and the ratio of under 20 µm or 5 µm becomes larger than that of the original soil. More specifically, coarse aggregates are crushed into slightly finer coarse particles and the surface is finely

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**Table 8** Change of weight and radioactivity fraction before and after MC for pebble mill and ultrasonic disperser for Soil AF.

| Processing | Soil fraction | Dry weight (g) | Weight fraction | Radioactivity concentration (Bq/kg) | Radioactivity (Bq) | Radioactivity fraction |
|------------|---------------|----------------|-----------------|-------------------------------------|--------------------|-----------------------|
| Without MC | >75 µm        | 1,252.3        | 64%             | 387                                 | 484                | 8%                    |
|            | 20–75 µm      | 134.1          | 7%              | 3,916                               | 525                | 9%                    |
|            | <20 µm        | 561.2          | 29%             | 8,804                               | 4,941              | 83%                   |
| Pebble mill | >75 µm        | 708.4          | 64%             | 732                                 | 519                | 15%                   |
|            | 20–75 µm      | 59.5           | 5%              | 3,809                               | 226                | 7%                    |
|            | <20 µm        | 333.8          | 30%             | 7,850                               | 2,621              | 78%                   |
| Ultrasonic disperser | >75 µm        | 457.1          | 64%             | 38                                  | 18                 | 1%                    |
|            | 20–75 µm      | 33.6           | 5%              | 4,160                               | 140                | 6%                    |
|            | <20 µm        | 220.2          | 31%             | 9,148                               | 2,014              | 93%                   |

**Table 9** Conditions for experiments.

| Item                      | Run 1 | Run 2 | Run 3 | Run 4 | Run 5 | Run 6 |
|---------------------------|-------|-------|-------|-------|-------|-------|
| Soil                      | Soil A (Kanuma Andisol) | Soil Y (Daigo soil) |
| Time for crushing (min)   | 10    | 40    | 40    | 16    | 15    | 16    |
| Rotation speed (rpm)      | 24    | 24    | 24    | 45    | 45    | 45    |
| Crushing media            | 10-mm ball | 10-mm ball | 6-mm ball | 5-mm Asake sand |
| pH                        | 7     | 7     | 7     | 7.7   | 11.5  | 11.5  |
| Immersion before MC       | -     | -     | -     | -     | -     | 10 min, 45 °C |

![Fig. 11 Changes in particle-size distribution due to MC.](image)
decomposed by friction. However, since it is not possible to determine whether or not only the organic aggregates bonded by humus were selectively decomposed only from the change in particle-size distribution, Figs. 12-14 should be jointly analyzed.

Figure 12 shows the carbon content for each particle-size fraction estimated by XRF, and the carbon distribution on a dry soil basis is calculated using the particle-size distribution and carbon content, which is shown in Fig. 13. In Fig. 12, despite the decrease in the amount of organic matter shown in Fig. 13, the high carbon content in the large-grained fraction after MC shown in red, green, and blue indicates the presence of relatively large particles of organic matter, such as wood chips and roots. Figure 13 suggests that both volume and surface decomposition occurred at the same time because the distribution of organic matter shifted to the fine grain side (in the order of red, green, and blue from original soil) due to MC. Also, generally speaking, the longer the MC time and the smaller the ball diameter, this tendency became more pronounced.

Figure 14 is a replot of Fig. 12 and is a simplified version of the carbon content for each particle-size fraction before and after MC with 6-mm balls. Comparing the 6-mm balls (indicated by blue bars), which are considered to be the most decomposed, with the original soil, it was found that the proportion of organic matter decreased in the range of 38-710 µm, but increased with finer fractions, which indicates that the clay with organics adhering to the surface of the coarse soil particles was stripped off.

These results confirmed that the decomposition of the target soil effectively progresses while both the volume and surface decomposition shown in Fig. 2 occur by the pebble milling using a medium measuring several millimeters, and that selective decomposition of only organic aggregates with low adhesive force without destroying sand and gravel is successfully achieved. Regarding the MC time, it is clear that a longer MC time increases the decomposition, but it is necessary to determine an appropriate time based on the target classification point and decomposition rate in the subsequent separation process.

Table 10 shows the particle size of each fraction measured by SALD-2300 before and after MC of the original soil and Runs 4-6 in volume percent. The fact that every fraction of 60, 30, and 10 µm or less increased confirmed that the decomposition progressed effectively for fractions larger than 60 µm in the Andisols. A slight increase in the fraction of Runs 4-6 below 60 µm indicates the possibility of the effect reaggregation prevention and the effect of immersion.
in warm water before MC.

(5) Examination of applicability to volume reduction and recycling of typical removed soil

First, the formulation to estimate how much soil can be recycled is examined by the soil size separation process considering the material balance of radioactive cesium and soil. Where $i$ is the number of fractions of the processing, $R$ is the radioactivity concentration of soil before treatment, $R_i$ is the radioactivity concentration of soil fraction $i$, and $M_i$ is the weight ratio of the treated soil of fraction $i$ whose total $M$ is equal to 1, the material balance of radioactive cesium and soil is expressed by equations (1) and (2).

\[ RM = \sum_{i} R_i M_i \quad (1) \]
\[ M = \sum_{i} M_i \quad (2) \]

Assuming that the decontamination rate of each fraction by the applied MC technique is $\alpha_i$, Eq.(1) can be transformed as in Eq.(1').

\[ RM = \sum_{i} (1 - \alpha_i) R_i M_i \quad (1') \]

In case $\bar{X}$ as the regulation value for recycling, it is sufficient to satisfy Eq. (3) for recycling. Note that $m$ is the last number of soil fraction $i$ that can not be recycled.

\[ \bar{X} \geq \sum_{i=1}^{m-1} (1 - \alpha_i) RM_i / \sum_{i=1}^{m-1} M_i \quad (3) \]

In the current separation process which takes only 75 $\mu$m as the classification point, Eq. (3) is expressed as Eq.(3a) with $i = 1$ for the particle size range of $>75$ $\mu$m and $i = 2$ for $\leq 75$ $\mu$m.

\[ \bar{X} \geq (1 - \alpha_1) R_1 \quad (3a) \]

Using the typical particle size distribution of the removed soil as in Table 11 and 8,000 Bq/kg for $\bar{X}$, $R_i$ becomes 32,000 Bq/kg or less from Eq.(3a) in case $\alpha_1 = 0.75$ that is a rough estimation of the current separation process. Eq.(4) is the relationship between the radioactivity concentration in year 2015 and the radioactivity concentration after 30 years for $^{137}$Cs neglecting $^{134}$Cs whose half-life is as short as 2 years.

\[ R = \left( \frac{1}{2} \right)^{\frac{2045-2015}{30.17}} R_{30} \quad (4) \]

From this equation, it is calculated that removed soil up to 63,751 Bq/kg as of 2015 can be recycled for the soil particle fraction of $>75\mu$m.

In the proposed FPSSS, 75$\mu$m and 20$\mu$m are the classification points, putting $i$ from 1 to 3 as shown in Table 11. Note that $i = 3$ is a fraction that is not recycled and is sent to other treatment, such as heat treatment, if recycle is required. Eq. (3) can be written as follows:

\[ \bar{X} \geq \frac{(1 - \alpha_1) RM_1 + (1 - \alpha_2) RM_2}{M_1 + M_2} \quad (3b) \]

Assuming that the decontamination rate of $>75$ $\mu$m fraction is 90% whose weight fraction is 50% and the decontamination rate of the 20-75 $\mu$m fraction is 50% whose weight fraction is 15%, removed soil with the radioactivity concentration up to 41,600 Bq/kg can be recycled. Converting to the radioactivity concentration as of 2015 from Eq. (4), the maximum radioactivity for recycling is calculated as 82,876 Bq/kg. The recycling rate at this time is 65% compared to 50% for the case of current soil size separation.

Second, from the results obtained in this study, applicability to the volume reduction and recycling of the removed soil is examined using the formula above. Figure 15 shows the amount of removed soil in Fukushima surveyed by MOE as of 2015 for each radioactivity concentration level. According to MOE, the condition for recycling of the removed soil is set to be 8,000 Bq/kg or less. Considering the decay of radioactivity in 30 years, the amount of removed soil that requires some treatment for recycling is in

| Table 11 Notation of $i$ and fraction of soil particle size. |
|-----------------|-------|------|------|
| Soil size fraction | 1     | 2     | 3     |
| $M_i$            | 0.5   | 0.15  | 0.35  |

Fig.15 Estimated volume of sandy and clayey soil for $^{137}$Cs plus $^{134}$Cs radioactivity concentration.
the range of 20,000 Bq/kg (which becomes 8,000 Bq/kg in 30 years) to 80,000 Bq/kg, among which 54% is in the range of 20,000-30,000 Bq/kg and 79% is in the range of 20,000-40,000 Bq/kg as shown in Fig.15. From the results of Tables 6-7, the average decontamination rate(α₂) and the weight ratio(M₂) for the 20-75 μm fraction of Soil N and Soil S is 25% and 5%, respectively, taking bebble mill as MC equipment. For the fraction of >75 μm, average decontamination rate(α₁) and the weight ratio(M₁) of Soil N and Soil S is 63% and 42%, respectively. With these values, removed soil with radioactivity concentration up to 19,492 Bq/kg can be recycled by proposed treatment, which is calculated as 38,832 Bq/kg as of 2015 from Eq. (4).

Then the amount of soil recycled is calculated to be 405,395 m³ from the volume profile of Fig. 15, which is an increase of 44,802 m³ from the standard particle-size separation of 75 μm or more. According to MOE, the cost of classification processing is in the order of 15,000 to 31,000 yen, and the cost of heat treatment is in the order of 100,000 to 220,000 JPY. Considering these costs, 11% increase in recycling rate with proposed system could be significant even though this shall be improved by further study.

5. CONCLUSION

In this study, the pebble mill and ultrasonic disperser were shown to be effective in a comparison of four types of MC equipment for the decomposition of soil from farmland aggregated due to the existence of organic matter containing humus. It was also shown that by combining MC and FPSSS, radioactive cesium can be concentrated in the fraction containing particles of 20 μm or less, making it possible to reuse soil particles >20 μm because the target radioactivity has been reduced. However, the results obtained in this study are 2%-39 % for the decontamination rate of the 20-75 μm fraction and 32%-65 % for the recovery rate of removed farmland-derived clayey-soil, and it is necessary to further improve the system and perform application tests on various soils.

In addition, through the pot-type pebble mill tests, basic characteristics, such as the change in particle-size distribution of soil particles and shift in the organic matter distribution, were confirmed. This shows that selective decomposition of organic aggregates that loosen only the bonds due to humus without destroying the particles themselves, such as sand, is possible.

In order to verify the effectiveness of the proposed system in the actual project, general formulas were prepared to calculate the radioactivity concentration that satisfies the regulation value for recycling from the radioactivity concentration of the raw soil, the decontamination rate, and the weight ratio of each particle-size fraction assuming the MC mechanism does not change significantly. Applying the results obtained in this study, it was found that the amount of recyclable soil increased by about 11%, which turned out to be significant considering the amount of removed soil and the treatment cost. Since MC used in this study has decontamination effect not only for the fraction of 20-75μm but also for the fraction of >75 μm, it might be possible that decontamination rate of the fraction of >75 μm is much larger if proposed MC is applied to this fraction, too.

However, even though this research aimed to increase the amount of recyclable soil by the combination of MC and particle-size classification, as the decomposition progresses, more soil particles are transferred to the finer fractions. With this treatment, the radioactivity concentration in the upper fraction becomes lower; but conversely, the amount of soil that can be recycled is reduced. At present, the Japan Ministry of Environment allows the reuse of soil containing 8,000 Bq/kg or less, and to increase the amount of recyclable soil meeting this requirement, the MC time should be adjusted according to the radioactivity concentration in the original soil. From the viewpoint of reducing the radioactivity concentration and improving safety as much as possible, a long MC time can be considered preferable. Hence, the required performance of the total system, from decontamination to recycling of contaminated soil, should be clarified and then optimized accordingly.

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REFERENCES
1) Ministry of the Environment, Japan: Strategy for developing technology for volume reduction and recycling of removed soil at interim storage facility - Review to achieve strategic goals, 2019. (in Japanese)
2) Ito, K., Miyahara, H, Ujie, T., Takeshima, T., Yokoyama, S, Nakata, K., Nanano, T., Sato, T., Hatta, T. and Yamada, H. : Practical approach to decontamination of radioactive cesium-contaminated matter in agricultural region by improved wet classification and use of geomaterials, J. of the Atomic Energy Society of Japan, Vol. 11, No. 4, pp. 255-271, 2012. (in Japanese)
3) Toyohara, H. and Sato, A. : Cleaning and volume reduction of contaminated soil containing radioactive substances, J. Bioscience and Bioengineering, Vol. 92, No. 5, 2014. (in Japanese)
4) Ide, K., Miura, T., Jintoku, T. and Takada, N. : Classification washing treatment of radioactive cesium-contaminated soil, Report of Obayashi Corporation Technical Research
5) Mouri, M., Baba, N., Tsuchida, M. and Nakajima, T. : Mass-balance analysis and performance evaluation of radiocesium contaminated soil washing, J. Japan Society of Civil Engineering, Vol. 71, No. 4, pp. 112-124, 2015. (in Japanese)

6) Yamaguchi, N., Takata, Y., Hayashi, K., Ishikawa, S., Kuramata, M., Eguchi, S., Yoshikawa, S., Sakaguchi, A., Asada, K., Wagai, R., Makino, T., Akahane, I. and Hiradate, S. : Behavior of radiocesium in soil-plant systems and its controlling factor, Bulletin of National Institute for Agro-Environmental Sciences, No. 31, pp. 75-129, 2012. (in Japanese)

7) Committee for the review of the behavior of radioactive cesium in soil, Japanese Geotechnical Society: Review of the behavior of radioactive cesium in soil, 2015. (in Japanese)

8) Fujita, T., Wang, L. P., Yabui, K., Dodbiba, G., Okawa, K., Matsuo, S. and Nomura, K. : Adsorption of cesium ion on various clay minerals and remediation of cesium-contaminated soil in Japan, Resources Processing, Vol. 60, pp. 13-17, 2013.

9) Ministry of the Environment, Japan: Current status and future plan of technology demonstration project for volume reduction and recycling of removed soil -Okuma pilot project-, 11th meeting of technology development strategy study group for volume reduction and recycling of removed soil at interim storage facility, 19th Dec., 2019. (in Japanese)

10) Miura, K., Mamiya, T., Tsujimoto, H., Kawano, M. and Kusaka, E. : Improvement of particle-size separation method for cesium-contaminated clayey soil and its challenges, J. Society for Remediation of Radioactive Contamination in the Environment, Vol. 9, No. 2, pp. 59-68, 2021.

11) Makoto, K. and Masami, N. : Introduction to Soil Science, 2nd Edition, pp. 103-104, Buneidou, 2019. (in Japanese)

12) Stevenson, F. J. : Humus Chemistry, John Wiley & Sons, pp. 496, 1994.

13) Caravaca, F., Lax, A. and Albaladejo, J. : Aggregate stability and carbon characteristics of particle-size fractions in cultivated and forested soils of semi-arid Spain, Soil & Tillage Research, Vol. 78, pp. 83-90, 2004.

14) Asano, M. and Wagai, R. : Evidence of aggregate hierarchy at micro- to submicron scales in an allophanic Andisol, Geoderma, Vol. 216, pp. 62-74, 2014.