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POSSIBILITY OF NEUTRAL-BASED SOLIDIFYING MATERIALS ON PREVENTING ELUTION OF RADIOACTIVE SUBSTANCES FROM RESERVOIRS

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ABSTRACT: The radioactive substances diffused with the occurrence of the 2011 Fukushima I Nuclear Power Plant accident is still deposited in the sediment of the reservoir. In this research, based on the dynamics of radioactive substances in soil by previous research, focusing on solidification of the reservoir sediment, the authors examined the applicability of neutral-based solidifying materials to be used at that time. In particular, the authors compare the difference between cement-based solidifying material and two types of neutral-based solidifying materials and discuss whether it is desirable to use any solidifying material in solidification of the reservoir sediment from the environmental impact after the improvement. In addition, the authors conducted a test on the characteristics required when using neutral-based solidifying materials.

Keywords: Cone index, Neutral-based solidifying materials, pH, Radioactive substances, Reservoir

1. INTRODUCTION

Radioactive substances released into the atmosphere due to the accident of Tokyo Electric Power Company Fukushima I Nuclear Power Plant generated by Tohoku Pacific Ocean Earthquake in 2011 was deposited on the ground surface over a wide range. Regarding the radioactive substances released by the accident, there are four substances which are the main problems in terms of health and environmental influences, iodine, cesium 134, cesium 137 and strontium. Various other substances have been released, but it is known that both have short half-lives or small radioactivity as compared with these 4 types [1]. For example, although iodine has a short half-life of 8 days, when entering the body 10 to 30% accumulate in the thyroid gland. Then, the thyroid gland receives exposure to beta rays and gamma rays for a while. In the case of Fukushima I Nuclear Power Plant accident contamination, there are two types of radioactive cesium, cesium 134 and cesium 137. Cesium 137 has a long half-life of 30 years, and environmental pollution lasts for a long time. Because radioactive cesium is similar in chemical nature to potassium, when entering the body, it is distributed almost entirely like potassium. Because strontium has a long half-life and chemical properties resemble calcium, it accumulates in bone when entering the body. Also, because strontium does not emit gamma rays, it cannot exactly find out where cesium 134 and 137 easily is. In case of an accident of Fukushima I Nuclear Power Plant, it is thought that strontium generated by fission is also present though it is less in quantity than cesium 134 and 137. Plutonium and others derived from the Fukushima I Nuclear Power Plant accident have also been detected, but quantitatively it is comparable to the measurements observed nationwide before the accident occurred.

In order to evaluate the influence on the environment by released radioactive substances, investigations on accumulation and transfer of radioactive substances in forests and farmlands occupying a large area immediately after the accident have been carried out continuously [2], [3]. At the same time, empirical studies on the actual condition of contamination by radioactive substances and decontamination have been conducted, and many findings have been obtained while being compared with the research at the Chernobyl nuclear accident [4].

The decontamination stipulated in Japan has priority over decontamination of agricultural land, but there is a possibility that agricultural land will be re-contaminated with radioactive substances via agricultural water supplied from a reservoir. However, decontamination of the reservoir has been postponed. This is because the contaminated soil in the reservoir was not regarded as a decontamination waste by Pollution Deal Special Measures Law for radioactive substances, and the accepting facility was not decided for a long time. In addition, the concentration of dissolved cesium in the environmental water such as groundwater, river water, reservoir and the like at normal times was almost within the standard, so urgency was not observed in decontamination of reservoir [5].

However, the policy was changed in FY 2014, and the Ministry of the Environment and the Ministry of Agriculture, Forestry and Fisheries announced to implement countermeasure on radioactive substances at reservoir [6]. The flow of countermeasures against radioactive substances at the reservoir is shown in Fig. 1. Regarding reservoir for agriculture, even though it is not a reservoir (see Fig. 2) meeting the criteria of decontamination conducted by the Ministry of the Environment based on Pollution Deal Special Measures Law for radioactive substances, measures must be taken from the viewpoint of resuming farming and reconstruction of agriculture under the technical support of the Ministry of Agriculture, Forestry and Fisheries, prefectural and municipalities were able to take countermeasure projects with the Fukushima Revitalization Acceleration Grant [7].

The decontamination method of the reservoir is selected according to the purpose and various conditions by the Technical Measures Manual for radioactive substances at reservoir prescribed by the Ministry of Agriculture, Forestry and Fisheries [8]. Conventionally, many methods of removing sediment contaminated with radioactive substances have been adopted, but there is a limit to carrying out the removed contaminated sediment and storing it in a prescribed storage facility. Therefore, in recent years the necessity to take measures by solidification of the sediment according to reservoir scale and pollution situation is increasing. Therefore, it can be expected that the number of construction projects by solidification of the sediment will increase. In the decontamination method by solidification of the sediment, a ground improvement material is used to harden the sediment contaminated with radioactive substances. Generally, there are many construction examples where the cement-based solidifying material is used. Previous studies have shown that it is possible to prevent the outflow of radioactive substances by using cement-based solidifying material [9]. However, the sediment improved with cement-based solidifying material shows a strong alkalinity with pH 11-12. For this reason, alkaline components are eluted from the sediment of the surface, and there is a problem that the water quality of reservoir exhibits alkaline properties. It is also demonstrated that radioactive substances tend to be eluted more easily as the soil is in the acidic region [10].

This study considers that it is effective to keep the pH concentration of the target soil in the neutral zone when confining the radioactive substances of the reservoir sediment and focuses on the neutral-based solidifying materials which do not change pH of the soil. Furthermore, using the neutral-based solidifying materials, the authors verify the possibility of preventing outflow of radioactive substances from the reservoir by solidifying reservoir sediment.

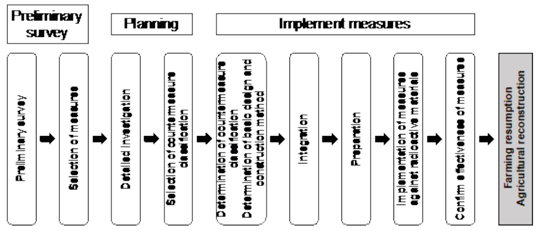

Fig. 1 Flow of radioactive material countermeasure for reservoirs

Fig. 2 Classification of jurisdiction over reservoirs

2. CHARACTERISTICS OF NEUTRAL-BASED SOLIDIFYING MATERIALS

2.1 Neutral-based Solidifying Materials

Hemihydrate gypsum or magnesium oxide is often used as the main ingredient of the material called current neutral-based solidifying materials [11] and solidification is carried out without changing the pH of the target soil. Consequently, the concern of changing the pH of the target soil by the cement-based solidifying material or the lime-based solidifying material to alkaline and adversely affecting the surrounding environment can be solved by using neutral-based solidifying materials. However, the strength characteristics of the treated soil using the neutral-based solidifying materials are inferior to the case of using cement-based or lime-based solidifying material in terms of strength development [12]. Therefore, if neutral-based solidifying materials are required to have a certain strength, it is one of the tasks that additive amount is much larger than cement-based and lime-based types. In addition, it has been shown in the previous study [13] that cement-based solidifying material can prevent elution of radioactive cesium, but the possibility of neutral-based solidifying materials has not been completed yet.

In this research, the authors focus on the characteristics of neutral-based solidifying materials and evaluate the possibility of preventing radioactive substances outflow by solidifying sediment of reservoir contaminated with radioactive substances using neutral-based solidifying materials.

The neutral-based solidifying materials used in this study is a paper sludge based solidifying material and a gypsum-based solidifying material. The properties of each material are shown below.

2.1.1 Paper sludge-based solidifying material

(1) Because no cement is used, there is no worry of hexavalent chromium elution.
(2) The soils with high water content such as dredging of the reservoir can be improved.
(3) Immediately after mixing, it can be quickly improved by agglomerating without exotherm.
(4) It is an inorganic powder which can contain odor and harmful substances and shows a neutral zone when the reaction is completed.
(5) Lead contamination soil can suppress elution by mixing Paper sludge-based solidifying material.
(6) The improved soil is excellent in permeability and water retention, rich infusion with existing soil, promotes vegetation.
(7) Improved soil does not re-elute / resuspend even on returning to the surface of the soil, the ocean floor or the lake bottom.

2.1.2 Gypsum-based solidifying material

(1) Because the solidification is fast, the solidification process of the soils with high water content can be continued and the treatment cost can be reduced
(2) Simultaneous solidification and granulation, moderate to soil suitable for recycling by giving treated soil water permeability and water retention appropriate.
(3) Because the neutral-based solidifying materials do not change the pH of the soils with high water content by solidification, the treated soil is reformed into soil suitable for vegetation without hindering the vegetation held by the soils with high water content.
(4) Because treated soil has no fish toxicity, it does not harm the ecosystem in water.

2.1 Mechanism of Solidification

Neutral-based solidifying materials rapidly absorbs moisture and promotes agglomeration when mixed and stirred with high water content sludge and soils with high water content. As a result, it is possible to convert soils with high water content such as construction sludge and dredged sludge into the high-quality soil which is excellent in water permeability and not soils with high water content and to give strength to the soil by rolling.

Paper sludge-based solidifying material uses silica powder having a large crystal surface area and higher water absorption/adsorption performance as the main raw material. Silica powder refers to powders from which hazardous substances have been removed by processing paper sludge ash from paper mills. Ettringite (acicular crystals) are formed by reaction with natural minerals in the auxiliary additives, and heavy metals and the like can be confined and insolubilized. In addition, magnesium oxide which is a component of paper sludge-based solidifying material hydrates with water in the soil and produces magnesium hydroxide hydrate.

$$MgO + (n+2)H_2O \rightarrow Mg(OH)_2 \cdot nH_2O \qquad (1)$$

The produced magnesium hydroxide hydrate reacts with the phosphoric acid in soil and carbon dioxide gas in the atmosphere to become magnesium phosphate or basic magnesium carbonate, which increases the solidification strength.

$$5Mg(OH)_2 \cdot (n\text{-}4)H_2O + 4CO_2 \rightarrow 4MgCO_3 \cdot Mg(OH)_2 \cdot nH_2O \qquad (2)$$

Therefore, it is considered that paper sludge-based solidifying material can suppress the change to alkalinity by the buffering action of magnesium.

The gypsum-based solidifying material is a product obtained by pulverizing raw material dihydrate gypsum (CaSO4·2H2O) and calcining it into hemihydrate gypsum (CaSO4·1/2H2O). The calcined hemihydrate gypsum mixes and stirs with water so that it becomes hydrated gypsum again by hydration reaction within a short time, and the target soil can be solidified by curing at that time.

$$CaSO_4 \cdot 2H_2O \Leftrightarrow CaSO_4 \cdot 1/2H_2O + 3/2H_2O \qquad (3)$$

In addition, the gypsum-based solidifying material does not change the influence on the surrounding environment compared to cement-based solidifying material and so on. because the solidification material itself is neutral and the solidification reaction is completed in the neutral state. Also, because the calorific value during the reaction is small, there is no fear of ignition or burn due to heat generation.

While the gypsum-based solidifying material has such advantages, natural gypsum is hardly produced in Japan, so when producing gypsum-based solidifying material, it is generally preferable to use by-product chemical gypsum (flue gas desulfurization gypsum, phosphate gypsum, hydrofluoric acid gypsum and so on) is used as a raw material. However, major chemical gypsum contains fluorine compounds, so there is concern that treated soils constructed from solidifying materials made from them will create new Brownfield land. Therefore, the authors must think about countermeasures in the problem of fluoride elution in the gypsum-based solidifying material. Fluorine elutes unless fluoride insolubilized ($CaHPO_4 \cdot 2H_2O$) is added to gypsum-containing fluorine. In order to solve this problem, manufacturers of gypsum-based solidifying materials have developed high-performance fluorinated insolubilizers and have taken all possible measures to reduce fluoride elution of gypsum-based solidifying materials. Therefore, it can be safely used against environmental standards for contaminated soils. As shown in Fig. 3, the insolubilizer added as a measure for preventing fluoride elution can be reduced to fluoroapatite by chemical reaction and the elution of fluorine in the treated soil can be reduced to less than the reference value. In addition, the insolubilizer to be added is a calcium phosphate salt ($Ca_{10}(PO_4)_6F_2$) whose main component also exists in vivo, it is a safe substance and the mechanism of its insolubilization is clear.

2.2 Difference from General Solidifying Materials

The origin of the technical term “solidifying material” is cement-based solidifying material and lime-based solidifying material produced for ground improvement. Therefore, it is not a technical term born from the idea of hardening soil in the neutral zone. Most neutral-based solidifying materials express aggregation effect as solidification. Therefore, it is inferior to cement-based solidifying materials and the like in strength development and economy for ensuring trafficability of soft soil and for building up to the foundation soil, so cases used from special site situations. However, as above mentioned, the neutral-based solidifying materials have superior characteristics with respect to decontamination of reservoir compared to cement-based solidifying material and the like. In addition to these characteristics, in this research, the authors conducted a test to quantitatively understand the solidification properties and pH characteristics of neutral-based solidifying materials.

3. EXPERIMENTAL EVALUATION OF POSSIBILITY OF NEUTRAL-BASED SOLIDIFYING MATERIALS

The test objects are two types of neutral-based solidifying materials: paper sludge-based solidifying material and gypsum-based solidifying material. The component composition of paper sludge-based solidifying material and the gypsum-based solidifying material is shown in Figs. 4 and 5.

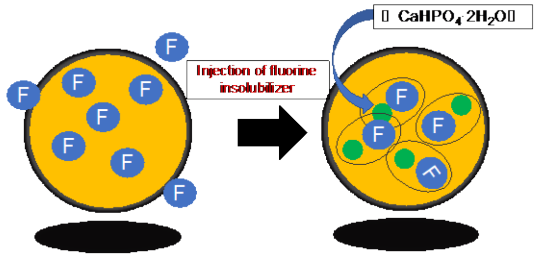

Fig. 3 Measures to prevent dissolution of fluorine

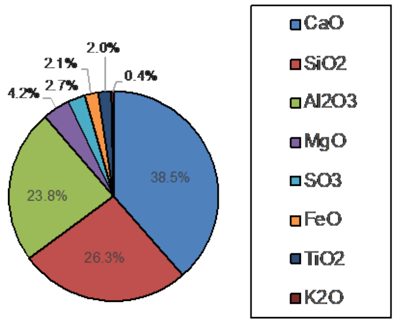

Fig. 4 Composition of paper sludge-based solidifying material

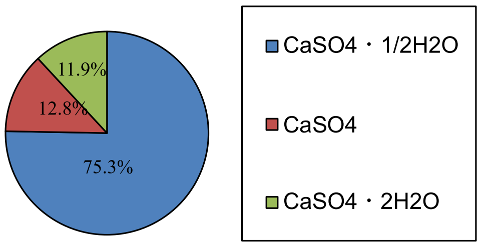

Fig. 5 Composition of gypsum-based solidifying material

Table 1 Properties of target soil

Target soil	Funabashi-silt
Water content (%)	58.2
Wet density (g/cm^3)	1.462
Dry density (g/cm^3)	0.924

In addition, quicklime and cement-based solidifying material are also used for comparison in the pH test.

For the test, conduct the cone index test of the soil and the pH test of the soil suspension. Each test method is shown below.

3.1 Test Methods

3.1.1 Cone index test

The cone index is the penetration resistance when pushing the corn penetrometer into the soil. Under the background of the “Law for the Promotion of the Use of Renewable Resources” which came into effect in October 1990, it is required to classify and use the construction soil according to the particle size and strength characteristics, and cone index is an index for this classification. Table 1 shows the amount of soil used for the cone index test. The same soil was used for the target soil of the specimen used for the pH test. The target soil used for the specimen is a Funabashi-silt having a unit volume weight of 1.6 kN/m^3 and a water content of 58.2%. In the cone index test, a comparison was made to the paper sludge-based solidifying material and gypsum-based solidifying material according to “Cone Index Test Method of Compacted Soil (JIS A 1288)”. The added amount of paper sludge-based solidifying material and gypsum-based solidifying material was 15% of the weight ratio of each specimen. Four specimens with different curing time (1, 3, 24 and 48 hours) were prepared, penetration resistance of each penetration amount (5.0, 7.5 and 10.0 cm) was read from the scale, The average penetration resistance was calculated and divided by the value of the cone bottom area value 3.24 cm^2 to determine the cone index of each.

3.1.2 pH test

Quicklime and cement-based solidifying material were also used. The target soil is Funabashi-silt used at the cone index test. 165 g of the target soil, 7% of the weight ratio of the specimen was added as the neutral-based solidifying materials and 200 kg/m^3 of the quicklime and the cement-based solidifying material were added, respectively. Because the authors assumed reservoir in this study, the pH concentration of each specimen was immersed in water with a pH meter at each elapsed time (1, 3, 24, 48 hours).

3.2 Results and Discussions

3.2.1 Cone index test

The results of the cone index test are shown in Fig. 6. In the cone index test, the cone index did not change until 24 hours passed since the initial strength was developed, and the strength increased at the lapse of 48 hours. Therefore, although it is a short time until the initial strength is developed, it is necessary to have 48 hours or more before the final reaction is completed. Also, in this test result, the authors could not obtain sufficient cone index as improved soil. Furthermore, with the use of neutral-based solidifying materials, it is difficult to carry out with standard specification dump trucks and to secure traffic-ability for heavy machinery to move on after improvement. In order to develop strength only with a neutral-based solidifying material, it is necessary to further increase the amount to be added or to compact after mixing and stirring.

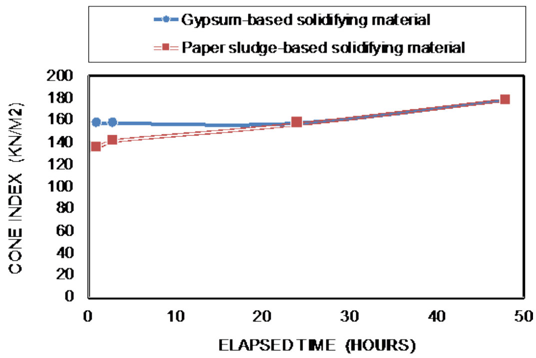

Fig. 6 Results of cone index test

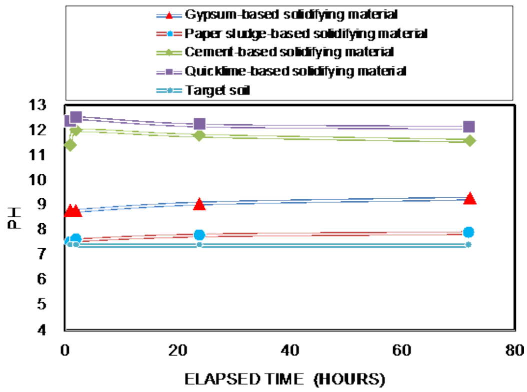

Fig. 7 Results of pH test

When the target soil is the same, the strength property of the treated soil using any neutral-based solidifying materials are significantly inferior in the ultimate strength development when compared with the case of using the cementitious or lime-based solidifying material. It is clarified in the previous study [14]. Therefore, when neutral-based solidifying materials are required to have a certain strength, the addition amount of neutral-based solidifying materials are considered to be much larger than that of cement and lime type.

In view of this result, the neutral-based solidifying materials are “a modifying agent with a large cohesive effect to be used when the treated soil itself must be neutral”, and “cement/lime type is a general variety solidification material that takes into account the increase in the ground tolerance of the ground so as to be used in the treatment plant”.

3.2.2 pH test

The results of the pH test are shown in Fig. 7. In the pH test, the pH value did not change substantially even after all of the four types of solidifying material were measured after 1 hour and after 72 hours. In addition, comparing pH of two types of neutral-based solidifying materials, paper sludge-based solidifying material showed higher pH value than gypsum-based solidifying material. This is thought to be the result of alkalization due to the alkaline nature of the silica powder which is the raw material of paper sludge-based solidifying material. In gypsum-based solidifying material, the authors did not change the pH of treated soil after solidification. This can be inferred that the treated soil after the reaction also maintains the neutral region because the main constituent and the product after the reaction show neutral. In addition, cement-based solidifying material and quicklime made it possible to confirm that the quality of the treated soil and the added water was changed to alkaline (pH 11 to 12).

Here the authors reconsider how soil bases that affect the pH value are present in the soil. In a very fine part of the soil, clay mineral and hazardous substances combine to form a clay/humus complex. Usually, this state is called soil colloid. As shown in Fig. 8, this soil colloid has a minus (-) charge on its surface and adsorbs cations such as calcium, magnesium, potassium and the like having positive (+) charge. Among the cations adsorbed and retained by this soil colloid and easily replacing other cations, those other than hydrogen ion are called exchangeable bases. Because exchangeable bases are most easily absorbed and used in agricultural crops, it is generally recognized that soil, which contains a large number of exchangeable bases in agriculture, is fertile. The hazardous substances mentioned above are those in which most of the organic substances entering soil as animal and vegetable bodies such as fallen leaves and falling branches are decomposed into water and carbon dioxide by the action of soil microorganisms and are collectively referred to as a part of remaining polymer compounds. Because the soil of this study assumes reservoir sediment, it is considered to exist in such soil state. In most soil environments, some of the acidic functional groups contained in the humus manifest negative charges by dissociating hydrogen ions, so humus can be an adsorbent of radioactive cesium.

Next, the relationship between exchangeable base and soil pH will be described. The soil has one showing acidity, one showing neutrality and one showing alkalinity. pH is used to express soil's reaction, that is, the degree of acidity or alkalinity, as a numerical value. The pH is called the hydrogen

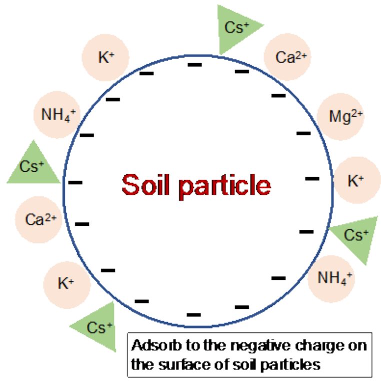

Fig. 8 Cations adsorbed to the soil particles

ion exponent and is defined as pH = -log[H^+], where [H^+] is the relationship of H^+ concentration in aqueous solution. The pH value in soil is one of the important properties, and the chemical form and solubility of the soil component change depending on the pH condition of the soil. In Japan with high rainfall, soil showing acidity is widely distributed. The reason why soil is acidic is the exchangeable aluminum ions and exchangeable hydrogen ions adsorbed and retained in acidic substances (nitric acid, sulfuric acid, carbonic acid, acetic acid) and soil colloids dissolved in the soil. When the minus charge of soil colloid is saturated with the exchangeable base, it is a neutral state but the base is leached by an external factor and hydrogen ion is adsorbed to soil colloid instead of the base to become acidic soil. It is known that radioactive substances already adsorbed on soil colloids are likely to be eluted as cations in such acidic soil [15].

In addition, as described above, the radioactive cesium once adsorbed on the sedimentation is mainly caught in the gap between the fine crystal structure of the tributary and the one bonded by the matching by the electrostatic force with the soil. It is thought that there is something missing [16].

The radioactive cesium being attracted to the soil by electrostatic force may release the radioactive cesium instead of attracting the chemical if it is exposed to chemicals with stronger force (ion exchange). However, because there are almost no chemicals existing closer to us than cesium (those with a larger ionic radius than cesium), there is a possibility that cesium once adsorbed on the sediment is re-eluted by ion exchange.

4. CONCLUSIONS

Environmental pollution caused by radioactive cesium is a serious problem, and decontamination in the reservoir is also essential. Because radioactive cesium has high water solubility, it releases a wide range of soil pollution through the atmosphere/hydrosphere circulation when released into the environment. For this reason, as a next step of the emergency decontamination which has been carried out until recently, it is an urgent matter to establish a technology that stably confines the radioactive cesium extracted and concentrated from the contaminated topsoil and prevent re-diffusion into the environment. In this research, the authors investigate the possibility of countermeasures against decontamination using neutral-based solidifying materials and summarize the findings of the neutral-based solidifying materials obtained below.

(1) The two types of neutral-based solidifying materials used in this study could not satisfy cone index 200 kN/m^2 for discharge as construction generated soil. Likewise, no strength development was observed to ensure traffic-ability to withstand construction heavy machinery. In addition, although two types of neutral-based solidifying materials used in this study can be completed in a short time until the initial strength is developed, it is necessary to have 48 hours or more before the final reaction ends It could be confirmed. Therefore, in order to develop strength only with neutral-based solidifying materials, it is necessary to further increase the amount added or to consider mixing the cement as a reinforcing material.

(2) It was confirmed that the gypsum-based solidifying material used in this study maintains the neutral region without changing the pH of the specimen. On the other hand, in paper sludge-based solidifying material, alkaline products are generated during solidification reaction, so pH is weakly alkaline. Further, in the pH test, it was confirmed that the pH value did not substantially change even when the measurement was carried out after 72 hours from the measurement of all four after 1 hour.

(3) Because neutral-based solidifying materials express the cohesive effect as solidification, it does not necessarily have excellent strength characteristics. That is, neutral-based solidifying materials are a modifying agent having a large cohesive effect to be used when the treated soil itself must be neutral. In addition, the cement/lime system is a solidifying material considering an increase in the ground tolerance of the ground, as it is used in various general processing workers.

Responding to countermeasures on radioactive substances at the reservoir, the countermeasure construction method is decided by comprehensively judging construction feasibility, effectiveness, economy and so on according to site conditions. In the actual construction practice, sediment removal is adopted, and there are very few cases where solidification of sediment is selected as a

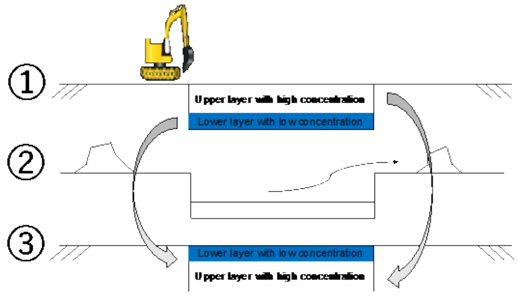

Fig. 9 Image of sediment reversal method

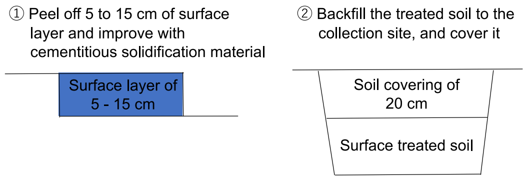

Fig. 10 Image of integrated reversal method

construction method. However, implementing the method shown in Figs. 9 and 10 and taking countermeasures, there is a possibility that countermeasures to contain radiation substances by sediment solidification are also possible. If the authors can prove that the neutral-based solidifying materials satisfy the cone index, it will be possible to pay attention to the water environment of the reservoir, so as a development of countermeasures against decontamination of the reservoir, the construction method using neutral-based solidifying materials also has size and degree It is necessary to use properly.

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