

OCEAN DECONTAMINATION: REMOVAL EFFICIENCY OF RADIOACTIVE CESIUM FROM OCEAN SLUDGE BY USING MICRO BUBBLES AND ACTIVATING MICROORGANISMS

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ABSTRACT: The Fukushima nuclear accident of March 11, 2011, soil and water had been contaminated by radioactive cesium. Moreover, radioactive cesium was found in the sludge in Tokyo Bay by flowing from rivers. Here, it cannot be easily removed the cesium which is adsorbed to the sludge. On the other hand, one of the authors had developed the decomposition system for sludge with circulation type by micro-bubbles and activating microorganisms. Here, based on the hypothesis that radioactive cesium is adsorbed on the surface of the sludge. It is considered radioactive cesium can be eluted, after decomposing the deposited sludge by the decomposition system. If the cesium will be eluted to the water, we can fix the cesium by existing technology such as “Zeolite”. In this study, our objects is to check the removal performance of cesium after the decomposition and elution from the sludge and then fix of cesium in the decomposition system. As the results, we obtained the removal efficiency of cesium was 62.7%.

Keywords: Decontamination, Radioactive Cesium, Ocean Sludge, Micro-bubble, Microorganism, Zeolite

1. INTRODUCTION

The Fukushima nuclear accident on March 11, 2011, soil and water had been contaminated by radioactive cesium. Moreover, radioactive cesium was found in the ocean sludge in Tokyo Bay by flowing from rivers. A report says it becomes 13 times of the cesium for 7 months from August, 2011 in [1]. Here, it cannot be easy to remove the cesium which is adsorbed to the sludge.

On the other hand, one of the authors had developed the decomposition system for ocean sludge with circulation type by micro-bubbles, which decompose and purification sludge by activating the aerobic bacteria, after creating an aerobic state by micro-bubbles.

Here, based on the hypothesis that radioactive cesium is adsorbed on the surface of the sludge deposition in [2], it is considered that radioactive cesium can be eluted, after decomposing the deposited sludge by using the decomposition system for ocean sludge with circulation type.

If the cesium will be eluted in the water, we can fix the cesium by the existing technology such as “Zeolite”.

In our past research in [3], removal performance of cesium was 30.9% by using the decomposition system for ocean sludge with circulation type by micro-bubbles and activating microorganisms. But Measurement of cesium by the iron chromatography was very sensitive so that we had to carry out the several experiments.

In this study, our objects is to check the removal performance of cesium after the decomposition and elution of the sludge and then fix of cesium by

setting “Zeolite” in the decomposition system for ocean sludge with circulation type by micro bubbles and activating microorganisms.

2. DECOMPOSITION SYSTEM WITH CIRCULATION TYPE

It is very important to reduce sedimentary sludge in the ocean. Plans to reduce the sludge are usually dredging or sand covering. Dredging is a simple way and aims to cut off the sludge. But after cutting off, treating the dredged sludge takes much more time and, of course, cost. Sand covering, in general, gives a big load to living organisms and the ecological system.

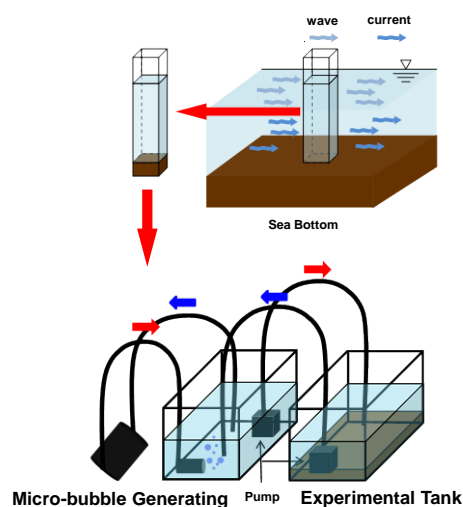


Fig. 1 Purification System of Circulation Type.

So that, a more efficient way is needed to reduce the sludge while not imparting environmental load in the local sea area. Here, attention was paid to micro-bubble technology for application to the purification of the sludge. The important point in this technique is to activate the bacteria existing in the area by micro-bubbles. Micro-bubbles (that is MB) can change conditions into an aerobic state. If the bubbling stops, the situation changes into anaerobic state, according to recent research. So, we selected a method for decomposing the sludge by microorganisms.

One of the authors had developed the decomposition system for ocean sludge with circulation type by micro-bubbles, shown in Fig.1, which decompose and purification sludge by activating the aerobic bacteria, after creating an aerobic state by micro-bubbles.

3. MECHANISM IN FIXING CESIUM FROM ELUTION

In general, ocean sludge has a negative charge. When cesium with a positive charge flows from river, sludge was adsorbed cesium, shown in Fig 2. So that, sludge adsorbed cesium cannot eliminate by usual way.

Here, we have a way by using of the decomposition system for ocean sludge with circulation type. After decomposition of the sludge adsorbed cesium by our system, cesium is eluted into water, shown in Fig 3. That is our hypothesis.

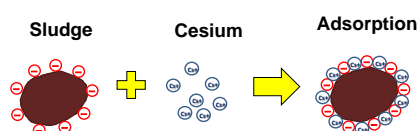


Fig.2 Mechanism on Adsorption of Cesium

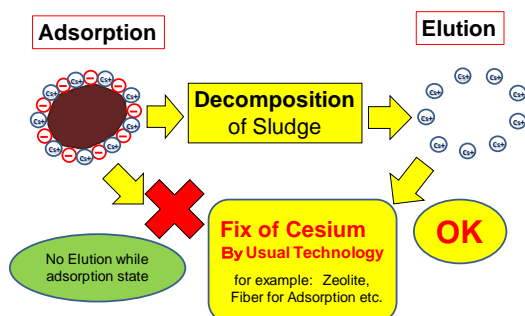


Fig.3 Mechanism on Fixing of Cesium from Elution.

4. REMOVAL EXPERIMENTS FOR CESIUM

4.1 Procedure of Experiments

The experimental devices consist of two parts, shown in Fig. 4. The water circulates through two tanks. In one tank (Width40xLength28xHight28cm), micro-bubbles are generated. The micro-bubbles have micro-size diameter and high solubility. This means the water with high concentration of dissolved oxygen circulates through these tanks. The other part is the experimental tank (W60xL29xH35cm). We used sea-water 30(litter) and sludge 1(kg). Here, a micro-bubble generator is based on [4], [5] and the flow rate is 900 (litter/hour). The flow rate of water pumps connected each tanks are 300 (litter/hour).

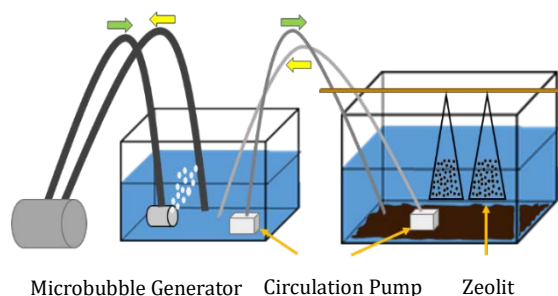


Fig.4 Experimental System for Elution of Cesium.

We had caught the sludge and the sea water at Funabashi Port in Chiba Prefecture in JAPAN, as shown in Fig.4 and 5. Here, we had removed under 10cm of the sludge from seabed before sampling as experimental procedure, because we have to remove the initial value of cesium in the sludge, from [3].

We used the cesium chloride before 24 hours of starting time and the concentration of cesium ion is 100 (ppm). A cooler for water tank was set at side of the tank for generating microbubbles, for the purpose of setting water temperature 30 degree centigrade.

After setting the decomposition system with circulation type by micro-bubbles, experiment starts at the same time of generating micro-bubble device and also the zeolites were set in the tank. After 6 hour, the microorganism activator was put in the experimental tank. Main staff of the activator is Kelp and including nutrients and some enzyme. Our used activator is reported to show effective results in purification for grease trap.

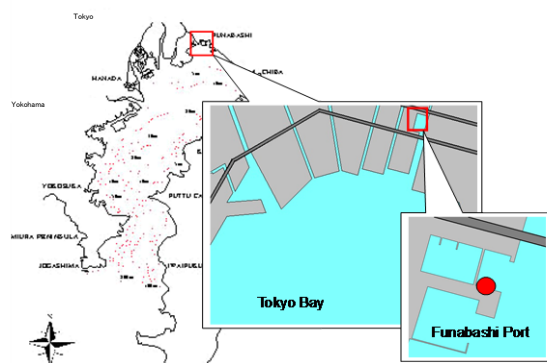


Fig.5 Catching Point of Sludge and Sea Water at Funabashi Port in Tokyo Bay.



Fig.6 Scene of Catching Sludge.

Dissolved oxygen (DO), water temperature and pH are measured by using of multi-parameter water quality meter. Ammonium nitrogen ($\text{NH}_4\text{-N}$), total nitrogen (T-N) and total phosphorus (T-P) are measured by using of digital-water-analyzer by digital "Pack test", by water filtered after sampling in experimental tank.

4.2 Experimental Conditions

Basically, zeolite have small and so many halls. As the results, it can fixed the cesium. We used the zeolite by composed type for experiment. Since the diameter of cesium is in general about 0.338(nm), we selected 4A type which has nearly diameter, and also 3A and 5A type of zeolite. And more selected Z13 type which has not uniform diameter, 0.2 to 1.0(nm). From the above, these are experimental conditions, shown in Table.1.

Table 1 Experimental Conditions

	Kinds of Zeolite
Case 1	3A ($\approx 0.3\text{nm}$)
Case 2	4A ($\approx 0.4\text{nm}$)
Case 3	5A ($\approx 0.5\text{nm}$)
Case 4	Z13 ($\approx 0.2\text{-}1.0\text{nm}$)

4.3 Results and Discussions

4.3.1 Results of water temperature, pH and DO as environmental condition

Fig.7-9 show the water temperature, pH and DO (Dissolved Oxygen) as the results of environmental condition of this experiment.

Water temperature is almost constant about 30 degree centigrade after 6 hours, by setting the cooler for water tank in the experimental system. pH is also constant about 7.5 to 8.7. The difference between Case 1, 2 and Case 3, 4 is caused by zeolite adsorb some ion when sludge is decomposed. DO in all cases is saturation state after 24 hours.

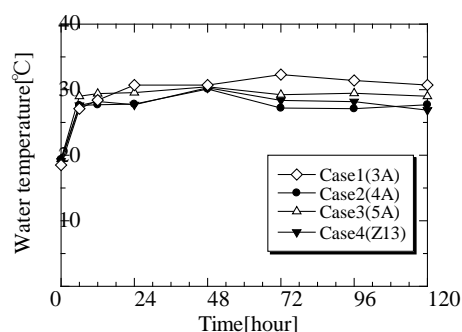


Fig.7 Changes in Water Temperature as Environmental Conditions.

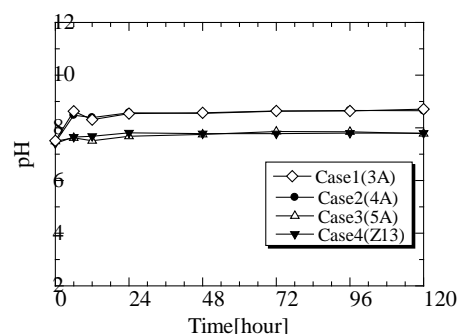


Fig.8 Changes in pH as Environmental Conditions.

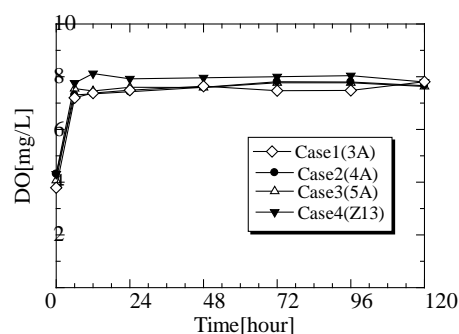


Fig.9 Changes in DO as Environmental Conditions

4.3.2 Results of H_2S , NH_4-N , DIN and T-N

Fig.10-13 are shown in results of H_2S (Hydrogen sulfide), NH_4-N (Ammonium nitrogen), DIN (Total inorganic nitrogen) and T-N (Total nitrogen).

H_2S in all cases decrease rapidly. It seemed by the supply of oxygen, and also another reason is zeolite can adsorb the H_2S . NH_4-N in all cases has tendency of decrease by 12 hours and then decrease slowly. DIN (= $NH_4-N+NO_2-N+NO_3-N$) shows 60% decrease. It seems it happen to denitrification by microorganisms from [7]. T-N in case 3 at 72 hours only increase. This reason is not clear but including measurement error. But, T-N in all cases decrease over 50% or maximum 75.0% by 120 hours. This purification efficiency is very good.

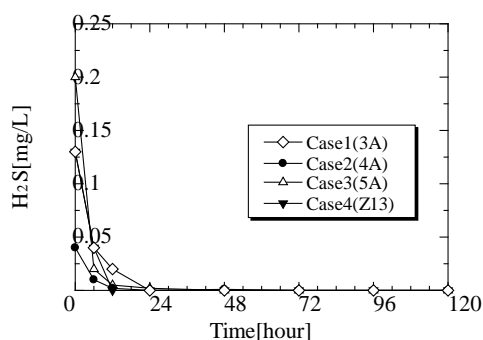


Fig.10 Changes in H_2S .

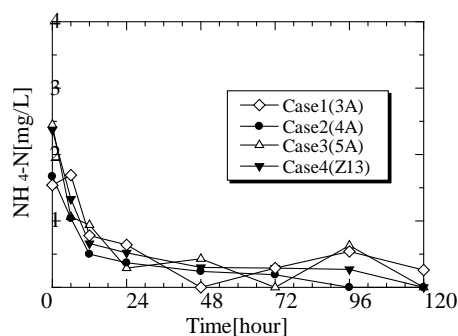


Fig.11 Changes in NH_4-N .

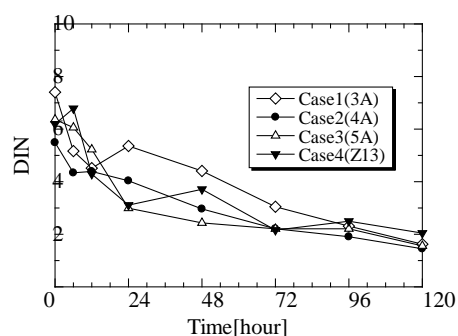


Fig.12 Changes in DIN.

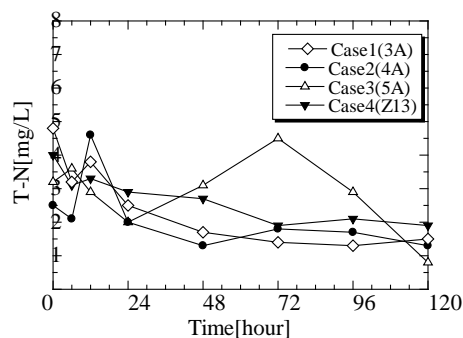


Fig.13 Changes in T-N.

4.3.3 Results of Cesium (liquid)

The results cesium were obtained by the iron chromatography shown in Fig 14.

The result of cesium is zero at 48 hours in case of 3A, and also is zero at 24 hours over in the other cases. It is especially very good the result of cesium in case 4A becomes zero until 12 hours. As the results of the above, fixed values of cesium by zeolite in all cases are 100 %.

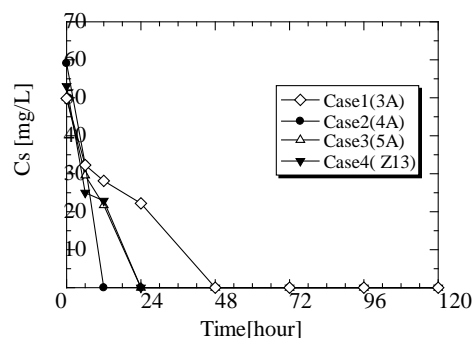


Fig.14 Changes in Cesium in Water.

4.3.4 Results of Cesium (solid)

We paid attention to cesium and silica (Si), as there are many chemical element in sludge. We used the energy dispersion type X-ray analysis device (EDX), because we can measure by the solid state. Weight ratio of cesium and silica (Cs/Si) in solid of dry sludge are shown in Fig.15.

Ratio of decontamination of cesium was calculated from ratio of content of beginning and final value measured by EDX, as the standard values including silica in dried sludge after measuring the weight of dried sludge.

Ratio of decontamination is obtained over 50% in all cases. Especially in case of 4A, the efficiency of decontamination is obtained 62.7%.

It seemed this experimental system including zeolite was obtained very good results and the mechanism on

fixing of cesium from elution by decomposition of the sludge.

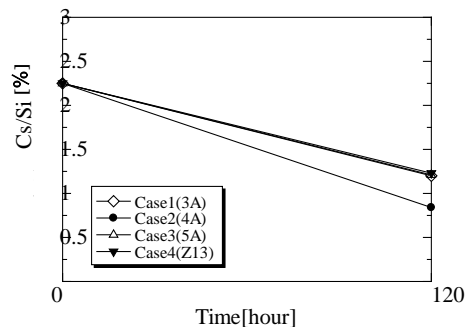


Fig.15 Changes in (Cesium)/ (Silica).

5. CONCLUSION

We had carried out the elution and fixing cesium by setting zeolite, after decomposing the deposited sludge, by using the decomposition system for ocean sludge with circulation type by micro-bubble and activating microorganisms. From the results by measurements for water qualification,

(1) T-N decreases maximum 75.0% in case of 3A. Purification efficiency is very good. From the results by iron chromatography,

(2) Fixed values of cesium by zeolite in all cases are 100 % and cesium in case 4A becomes zero until 12 hours. From the results of the energy dispersion type X-ray analysis device, the weight ratio of cesium and silica (CS/Si) in solid of dry sludge is denoted,

(3) The efficiency of decontamination is obtained over 62.7% in cases of 4A.

6. ACKNOWLEDGEMENTS

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7. REFERENCES

- [1] Yomiuri News Paper, 13 times by 7 months about cesium in ocean soil in Tokyo Bay, May, 13th, 2012.

- [2] Okamoto Kyoichi, Hotta Kenji, Toyama Takeshi and Kohno Hideki, Purification System of Ocean Sludge by Activating Microorganisms, International Journal of GEOMATE (Geotec, Const. Mat. & Env.), Vol.6, No.1, pp.791-795, March, 2014.
- [3] Okamoto Kyoichi and Toyama Takeshi: Ocean Decontamination: Removal of Radioactive Cesium from Ocean Sludge by Using Micro bubbles and Activating Microorganisms, The Japan Society of Naval Architects and Ocean Engineers, May, 2014.
- [4] Imai, T, Shiohige, K, Ukita, M, Sekine, M, Higuchi, T, Fukagawa, K and Fujisato, T. Development of Device for Dissolving High Concentration Gas for Purification at bottom layer around enclosed water area like Dam Lake, Proc. 39th Forum on Environmental Engineering of Civil Engineering Society, 10-12, 2002.
- [5] Matsuo, K, Maeda, K, Ohnari, H, Tsunami, Y, and Ohnari, H., "Water Purification of a Dam Lake Using Micro Bubble Technology", Progress in Multiphase Flow Research I, pp.279-286, 2006.
- [6] Okamoto Kyoichi and Hotta Kenji, Purification System of Ocean Sludge by Using Coagulants and Activating Microorganisms, International Journal of GEOMATE (Geotec, Const. Mat. & Env.), Vol.4, No.2, pp.574-579, June, 2013.
- [7] Sone Takaaki, Yamashita kazuhiko and Okamoto Kyoichi, Identification of Microorganism in Purification Process for Ocean Sludge by using Purification System with Circulation Type, Japan Association for Coastal Zone Studies, July 2014
- [8] Okamoto Kyoichi and Toyama Takeshi: Ocean Decontamination: Removal Performance of Radioactive Cesium from Ocean Sludge by Using Micro bubbles and Activating Microorganisms, The Japan Society of Naval Architects and Ocean Engineers, May, 2015.

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