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OCEAN DECONTAMINATION: HIGH ABILITY REMOVAL METHOD TO 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 ocean sludge in Tokyo Bay flowing from rivers. Cesium which is adsorbed to the sludge cannot be easily removed. One of the authors developed decomposition and purification system, a circulation-type system by micro bubbles, that is, by creating aerobic state, aerobic bacteria are activated resulting to decomposition and purification of ocean sludge. Based on the hypothesis that radioactive cesium is adsorbed on the surface of the sludge deposition. It is considered that cesium can be eluted after decomposing the deposited sludge. Once the cesium is eluted in the water, it can fix to a mineral such as zeolite. Eluting and fixing cesium adsorbed on sludge takes so much time. In this case, the concept of removing the left sludge by flocculation method and then followed by coagulating sedimentation method is studied. In this study, our objects is to show the effectivity and efficiency of using flocculation and coagulating sedimentation in removing radioactive cesium. As the results, we pointed out this method is very good.

Keywords: Ocean Decontamination, Radioactive Cesium, Micro Bubbles, Microorganism Activator, Coagulant

1. INTRODUCTION

The Fukushima nuclear accident of March 11, 2011, soil and water had been contaminated by radioactive cesium. Moreover, radioactive cesium was found in the ocean sludge in Tokyo Bay flowing from rivers. Cesium which is adsorbed to the sludge cannot be easily removed.[1] One of the authors developed decomposition and purification system, a circulation-type system by micro bubbles, that is, by creating aerobic state, aerobic bacteria are activated resulting to decomposition and purification of ocean sludge. [2]

Based on the hypothesis that radioactive cesium is adsorbed on the surface of the sludge deposition, it is considered that radioactive can be eluted after decomposing the deposited sludge. Once the cesium is eluted in the water, it can fix to a mineral such as zeolite. [3] But eluting and fixing cesium adsorbed on sludge takes so much time. In this case, the concept of removing the left sludge by flocculation method and then followed by coagulating sedimentation method is studied.

In this study, our object is to show the effectivity and efficiency of using flocculation and coagulating sedimentation in removing radioactive cesium.

2. HDECOMPOSITION SYSTEM WITH CIRCULATION TYPE

It is very important to reduce sedimentary sludge in the ocean. Plans to reduce the sludge are usually dreading 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.

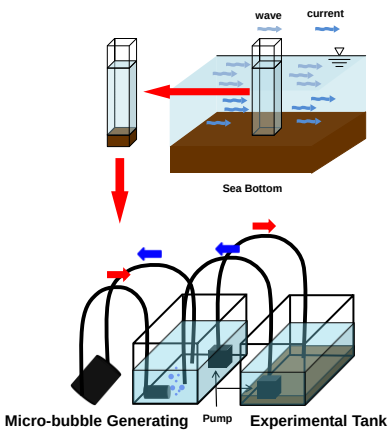

Fig. 1 Purification System of Circulation Type

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. [4], [5]

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]

3. MECHANISM ON REMOVING CESIUM

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.

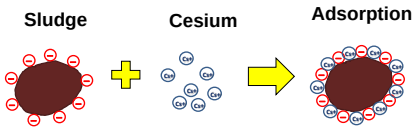

Fig. 2 Mechanism on Adsorption of Cesium.

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.

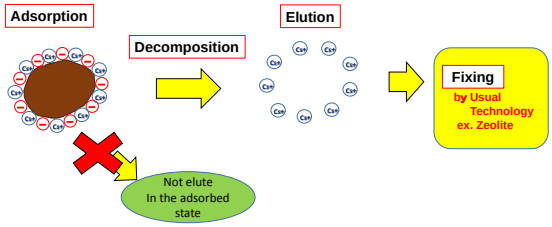

Fig. 3 Mechanism on Fixing of Cesium after Elution.

But, it takes much time to elute and fix all of cesium adsorbed on sludge. Now, we have new idea which is to use flocculation method against to the rest of decomposed sludge, and then make the precipitation of cesium and sludge, shown in Fig. 4.

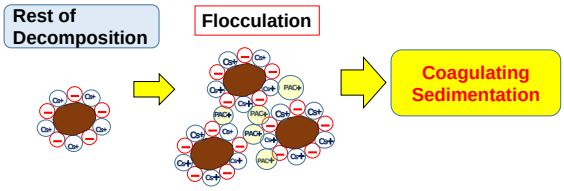

Fig. 4 Mechanism on Coagulating Sedimentation by PAC, to the left of Decomposition Sludge with Cesium.

4. REMOVAL EXPERIMENTS FOR CESIUM

4.1 Experimental System

The experimental devices consist of two parts, shown in Fig. 5. The water circulates through two tanks. In a tank (Width40xLength28x Hight28cm), 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). 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.

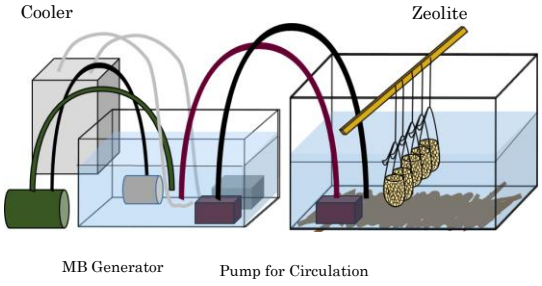

Fig. 5 Experimental System for Removal of Cesium.

4.2 Experimental Procedures

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 picked up the

sludge under 10cm from seabed before sampling as experimental procedure, because we have to cut 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).

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. The zeolite is the composed type and the pore size is 4A type that is 0.4[nm]. Diameter of cesium is generally 0.338[nm], so that this diameter is much closer.

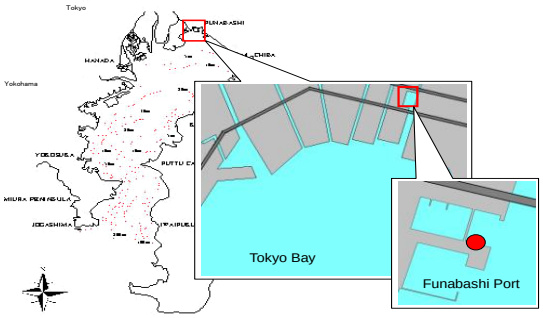

Fig. 6 Catching Point of Sludge and Sea Water at Funabashi Port in Tokyo Bay.

After 6 hours later, 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.

Dissolved oxygen (DO), water temperature and pH are measured by using of multi-parameter water quality meter. Ammonium nitrogen (NH_4-N), total nitrogen (T-N) are measured by using of digital-water-analyzer by digital "Packtest", by water filtered after sampling in experimental tank.

4.2.1 Procedure for Liquid Measurements

Measurements for liquid phase did each 6 hours until 12 hours, and then every 12 hours to120 hours, shown in Fig.7.

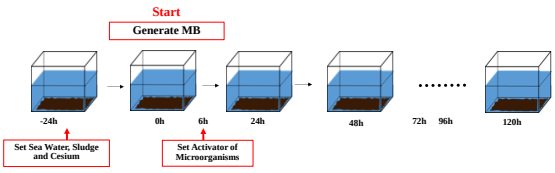

Fig. 7 Experimental Procedure for Liquid Measurements.

4.2.2 Procedure for Solid Measurements

As procedure of solid phase, experimental tanks are prepared for each measurement time; 0, 24, 48, 72, 96, 120 hours. When the objective tank after worked system is stopped the system, coagulant was put and mixing. After this, water quality and cesium are measured. After filtration and dry, cesium in solid was analyzed by the energy dispersion type X-ray analysis device (EDX). Here, our used coagulant is PAC (Poly Aluminum Chloride) and the solution concentration is 10%. [6]

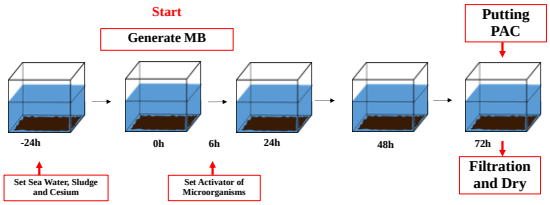

Fig.8 Experimental Procedure of for Solid Measurements (For Example: 72hours Case).

4.3 Experimental Conditions

Experimental conditions are the concentration of PAC according to the putting time, shown in Table 1. The another cases are PAC concentration is 500 ppm at 0, 24 hours, 25 ppm at 96 hours and 10 ppm at 120 hours.

Table 1 Experimental Conditions

at 48h	at 60h	at 72h
PAC (ppm)	PAC (ppm)	PAC (ppm)
600	200	100
400	100	75
200	75	50

5. RESULTS AND CONSIDERATION

5.1 Results on Water Quality

5.1.1 Water Temperature, pH, DO (Dissolved Oxygen) and H_2S (Hydrogen Sulfide)

As the results of environmental conditions; water temperature, pH and DO, water temperature was almost 30 centigrade degrees because of using water cooler. pH showed the values of 7.5 to 8.7. DO is saturation state by the values of 7.5 to 8.0. These results are almost the same as ordinary results. [3]

H_2S decreased rapidly by 24 hours by the supply of O_2 of microbubble device and then became zero at 48 hours. This is also the same as ordinary results. [3]

5.1.2 DIN (Dissolved Inorganic Nitrogen), T-N (Total Nitrogen) and Cesium in Water

DIN and T-N as purification items decreased well by the denitrification. T-N is reduced to 82%. This is also the same as ordinary results. [7], [8]

Cesium in liquid was analyzed by the iron chromatography, and the initial value for cesium is 50.06 (ppm) but the value by 12 hours is not detective, so that reduction rate of cesium was 100%.

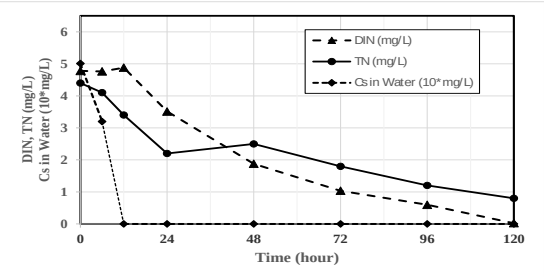

Fig. 9 Changes on DIN, TN and Cesium in Water.

5.2 Most Suitable Concentration of PAC

We are going to find the most suitable concentration of coagulant (PAC) depended on the working time, by putting the coagulant with concentration according to experimental condition.

By focusing on the T-N as the index of Table 2, the relationship of minimum concentration of PAC and working time of the system was shown in Fig.10. Here, the results of T-N were N.D. in case of 0, 24, 96 and 120 hours.

When it's the concentration of PAC of the upper part of a solid line in Fig.10, cesium of solid phase indicates the maximum purification performance

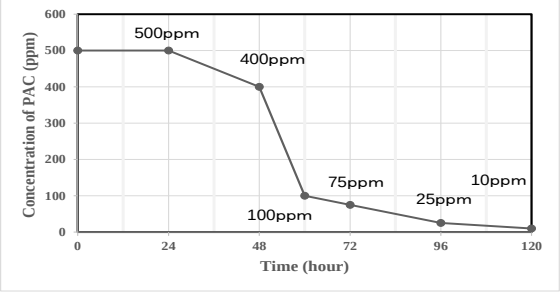

Fig. 10 Suitable Concentration of PAC According to Working Time in Experimental System

Table 2 Experimental Results.

at 48h		at 60h		at 72h	
PAC (ppm)	TN/TN0	PAC (ppm)	TN/TN0	PAC (ppm)	TN/TN0
600	ND	200	ND	100	ND
400	ND	100	ND	75	ND
200	0.24	75	0.22	50	0.20

and coagulating sedimentation is executed. Therefore, most suitable working time of the system is 48 to 60 hours, so that the reducing working time can be supposed and expected.

6. ADDITIONAL EXPERIMENT

We get the relationship of most suitable concentration of PAC according to working time of the system. But we need the changes of cesium before and after working system.

Now we executed the additional experiments for the purpose of getting of the amount of cesium before and after working system. The experiments are made in cases of 48 hours and 72 hours, and as comparative target is the case of 0 hour.

6.1 Water Quality

The results for water quality are shown Fig. 11. DIN and T-N are very good decrease by the denitrification. This is also the same as our ordinary results.

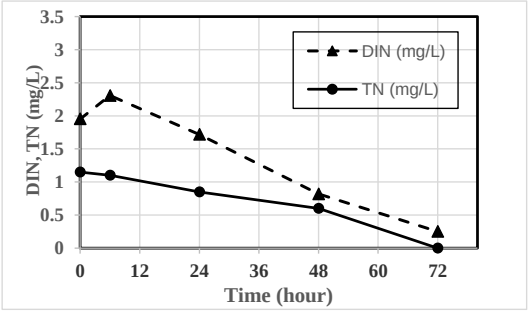

Fig. 11 Changes on DIN, TN in Additional Experiment.

6.2 Cesium in Water

Cesium in liquid was analyzed by atomic absorption spectrometry, and the value for cesium is not zero at 48 hours and 72 hours, shown in Fig.12. This is caused by weak circulation of experiment, and this analytical way is very good precision. But this is not big problem. We can understand the eluting cesium from sludge and have another way; adsorption cloth and other for cesium adsorption and fixing. The difference is small in the cesium amount before and after investment of PAC, in case of at 48 and 72 hours.

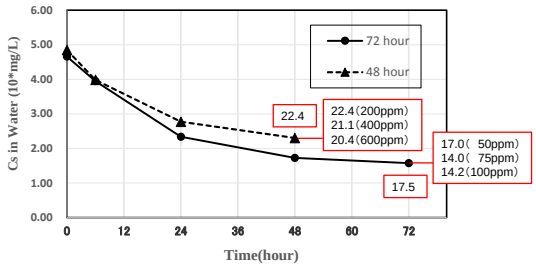

Fig. 12 Changes on Cesium in Water before and after investment of PAC.

6.3 About Safety to all Ecosystem by this System

Coagulant used in this experimental system is PAC (Poly Aluminum Chloride) and the solution concentration is 10%. [6] This is basically safe according to appropriate usage. It's supposed PAC is certainly the substance which isn't diffused around the environment. It seems it is no problem as far as clear layer at the top of liquid is discharged, because coagulant power is strong. Toxicity to short-necked clam and laver (Medial Tolerance Limit) is 10000(ppm)/48(hour) from safety data sheets (SDS). [9] Even if PAC we added doesn't have an influence on the environment. PAC hydrolyzes with passage in time and changes into a stable aluminum hydroxide.

7. CONCLUSION

We carried out the removing cesium in solid phase for experimental system which is to use flocculation method against to the rest of decomposition sludge, after the elution and fixing cesium by setting zeolite in the decomposition system with circulation type by micro-bubble and activating microorganisms.

(1) From the results by measurements for water qualification, T-N decreases maximum 82.0%. Cesium in water was fixed 100% and became zero by 12 hours of the experiment.
(2) We pointed out the relationship of most suitable concentration of PAC according to working time in experimental system. It assumed this concentration of PAC shows the maximum purification efficiency in water and is very good.
(3) It seemed the most suitable time for working the experimental system is 48 to 60 hours, so that the working time of system can be reduced.

We also executed the additional experiments for the purpose of getting of the amount of cesium before and after working system, in case of 48 hours and 72 hours, and 0 hour as comparative target. As the results are

(4) Cesium in water after stopping of the system and investment of PAC in case of 48 and 72 hours, is almost the same as the quality before investment of PAC.

8. ACKNOWLEDGEMENTS

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