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REMOVAL OF RADIOACTIVE CESIUM FROM OCEAN SLUDGE BY THE BACTERIUM USING PURIFICATION SYSTEM OF CIRCULATION TYPE

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ABSTRACT: The Fukushima nuclear accident of March 11, 2011, resulted in soil and water contamination by radioactive cesium. Radioactive cesium transported by rivers was also found in the ocean sludge of Tokyo Bay. Cesium adsorbed on the sludge is not easily removed. One of the authors has developed an ocean sludge decomposition system that employs micro-bubble circulation. The circulation of micro-bubbles creates an aerobic state that activates aerobic bacteria, facilitating decomposition and purification of the sludge. The objective of this study was to investigate the effect of the addition of bacteria to the micro-bubble circulation system on the efficient removal of radioactive cesium from ocean sludge. We tested the water purification effect of the system by adding bacteria directly. Results confirmed that the decomposition of the deposited sludge using our system facilitates the elution of the radioactive cesium. Any cesium eluted in the water can be remedied using existing technology such as zeolites. Purification efficiency seems to be greatest when additional bacteria are added directly to the process. With time, bacterial concentration doubles, and 76% of cesium in the liquid phase (dried sludge) and 51% of cesium in the solid phase (water) is removed.

Keywords: Radioactive Cesium, Ocean Sludge, Micro-bubble, Bacteria

1. INTRODUCTION

The 2011 accident at the Fukushima Daiichi nuclear power station led to radioactive cesium contamination of soil and water. Rivers flowing into the Tokyo Bay have transported radioactive cesium into the ocean sediments of the bay [1].

Cesium is reportedly easily adsorbed onto the microscopic particles that constitute soil [1-2]. Most cesium adsorbed by sediments is difficult to remove by external factors and persists over long timescales. Moreover, closed water environments such as river and bay systems make accumulated organic sludge difficult to decompose. Therefore, radioactive cesium is predicted to be deposited in the sediment of the seabed over time, extending radioactive contamination into the ocean. It is important to decontaminate the sediment.

Okamoto, one of the authors, developed a decomposition system for ocean sludge that employs the circulation of micro-bubbles to promote aerobic conditions that activate aerobic bacteria, facilitating decomposition and purification of the sludge [3]. Moreover, sea-sludge particles were observed to decrease in size through the course of the treatment with our micro-bubbles system and bacteria addition [4].

Radioactive cesium can thus be eluted after the deposited sludge is decomposed by our system. If the cesium is eluted into water, it can then be remedied using existing technology such as zeolites [5]; resulting in the complete decontamination of sediment.

In this study, to effectively remove radioactive cesium, we added bacteria to break down sludge in the micro-bubble circulation system and investigated the effect of the addition of microorganisms.

2. MATERIALS AND METHODS

2.1 Decomposition System of the Micro-Bubble Circulation Type

The micro-bubble circulation system comprises two parts, as shown in Fig. 1. The water circulates through two tanks. In one tank (length 40 × width 28 × height 28 cm), highly soluble, micro-bubbles are generated. Consequently, water rich in dissolved oxygen (DO) circulates through these tanks. The experimental tank is 60 × 29 × 35 cm. We used 30 L of seawater and 1 kg of sludge. The micro-bubble generator was based on [6] and the flow rate was 900 L/h. The flow rate of the water pumps connected to each tank was 300 L/h. A cooler for the tank that generates the micro-bubbles was set to 30 °C.

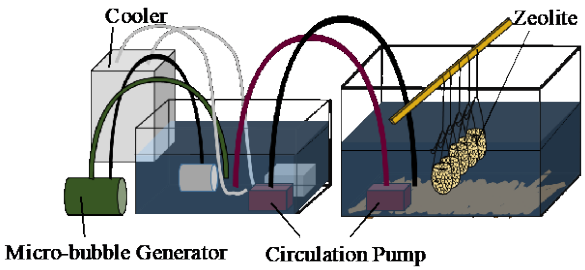

Fig. 1 Circulation purification system

2.2 Experiments on Decomposition of Deposited Sludge for Cesium Removal

Figure 1 illustrates the system used to experimentally remove cesium from ocean sludge. The ocean sludge samples and seawater were obtained from Funabashi Port in Chiba Prefecture, Japan. The first 10 cm of sludge from the seabed was discarded before the samples were taken. Model sludge was prepared using one kg of ocean-sludge, 30 L of seawater, cesium chloride was mixed. The cesium chloride was included to become 100 mg/L. The model sludge was poured into the system and stirred for 24 h. Zeolites 4A were placed in the second tank (Fig. 2), and micro-bubbles were generated by a micro-bubble generator using a flow rate of 900 L/h. The flow rate of the water pumps connected to each tank was 2400 L/h. The cooler was set at 30 °C.

After 24 h, bacterial species *"Alcaligenes fecalis"* (15×10^8 cells) [5,7,8] and Glutamic acid (100 ppm) as an activator [9] were added. The activator was added to the experimental tank after 24 and 60 h periods. At 0, 12, 24, 48, 60, 72, 96, and 120 h, DO, water temperature, and pH was measured by a multi-parameter water quality meter. A digital pack test (Kyoritsu Chemical-Check Lab. Corp., Japan) was used to measure ammonium ion (as ammonium nitrogen, NH_4-N), nitrite ion (as nitrite nitrogen, NO_2-N), nitrate ion (as nitrate nitrogen, NO_3-N), total nitrogen (T-N), and hydrogen sulfide (H_2S) concentrations. The seawater was then filtered.

Seawater and sludge samples at 0, 12, 24, 48, 72, 96, and 120 h were collected for DNA concentration and cesium concentration measurements. DNA was extracted from sludge and measured by spectrophotometry. Cesium was measured with energy dispersive X-ray analysis (EDX) in dried sludge and atomic absorption spectrometry in seawater.Experimental conditions are given in Table 1.

3. RESULTS AND DISCUSSION

3.1 Decomposition of Deposited Sludge for Cesium Removal

Fig. 2 Zeolites 4A set in experiment tank

Table 1 Experimental conditions

	Micro-bubble	Additive amount of bacteria	Activator
Case 1	○	1.5×10^8 cell	100 ppm
Case 2	—	–	–

3.1.1 Results of water temperature, pH, DO and H_2S under varying environmental conditions

Figures 3 and 4 show the pH and DO respectively, resulting from the environmental conditions prevalent in cases 1 and 2 (Table 1). After 24 hrs, the pH of case 1 and 2 was constant at about 8.0. The initial concentrations of DO are 4.9 mg/L and 3.7 mg/L in cases 1 and 2 respectively. The DO of case 1 is saturated at about 7.3 to 7.8 mg/L after 24 h, because the concentration of oxygen saturation is 8.1 mg/L-pure water. The DO of case 2 is saturated at about 3.5 to 4.2 mg/L after 24 h.

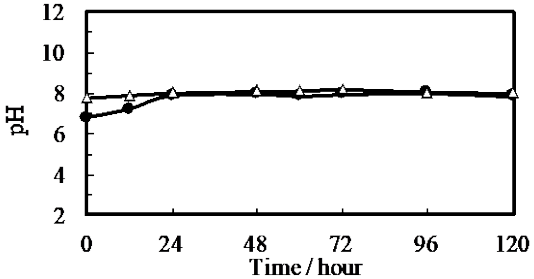

Fig. 3 Change in water pH over time in case 1 (●) and case 2 (Δ).

The H_2S concentration is shown in Fig. 5. In case 1, H_2S decreases up to 24 h; after which it is no longer detected, before the addition of *A. fecalis* to the experimental tank. *A. fecalis* is not inhibited

by H_2S. In case 2, H_2S decreased slightly and remained at 0.16 mg/L at 120 h.

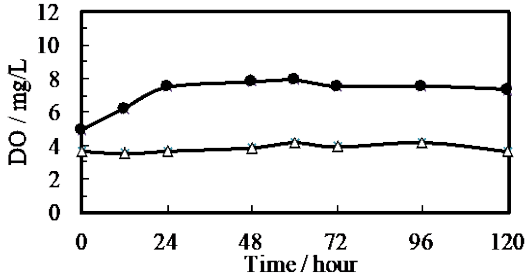

Fig. 4 Change in DO concentration over time in case 1 (●) and case 2 (Δ).

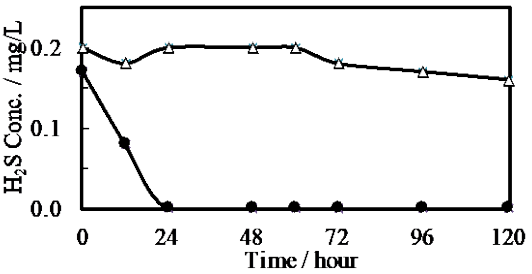

Fig. 5 Change in H_2S concentration over time in case 1 (●) and case 2 (Δ).

3.1.2 Results of NH_4-N, NO_2-N, NO_3-N, and T-N

Figure 6 shows the change in NH_4-N (ammonium nitrogen) concentration in the experimental tank over time. In both cases (Table 1), initial concentrations of NH_4-N are approximately 1.5 mg/L. In case 1, NH_4-N decreases and is no longer detected at 72 h. The effect of *A. fecalis* on NH_4-N concentration could not be ascertained because *A. fecalis* was added at 24 h. In case 2, NH_4-N increases to 1.8 mg/L at 120 h.

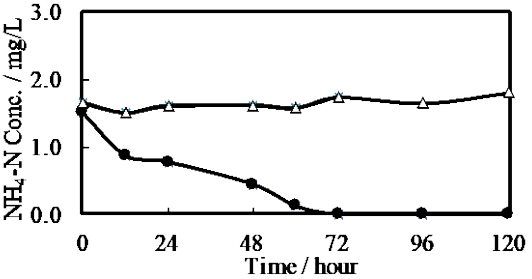

Fig. 6 Change in NH_4-N concentration over time in case 1 (●) and case 2 (Δ).

Figure 7 shows the change in NO_2-N (nitrite nitrogen) concentration in the experimental tank over time. In case 1, initial concentrations of NO_2-N are approximately 0.28 mg/L. Subsequently NO_2-N constant at 0.01 mg/L up to 48 h, after which it decreases and remained at 0.02 mg/L at 120 h. In case 2, initial concentrations of NO_2-N are approximately 0.28 mg/L. NO_2-N decreases to 0.13 mg/L at 48 h, then increases to 0.24 mg/L at 120 h.

Figure 8 shows the change in NO_3-N (nitrate nitrogen) concentration in the experimental tank over time. In case 1, initial concentrations of NO_3-N are approximately 3.7 mg/L. Following this, NO_3-N increases to 4.6 mg/L at 6 h, then decreases to 0.60 mg/L at 120 h.

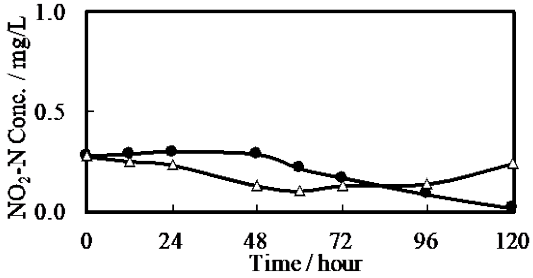

Fig. 7 Change in NO_2-N concentration over time in case 1 (●) and case 2 (Δ).

In case 2, initial concentrations are approximately 4.14 mg/L. Following this, NO_3-N increases to 4.7 mg/L at 24 h, then decreases to 2.2 mg/L at 96 h before continuing to gradually increase to 3.3 mg/L at 120 h

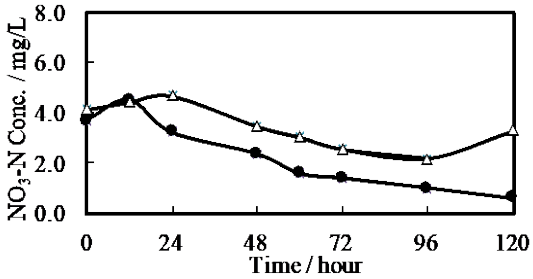

Fig. 8 Change in NO_3-N concentration over time in case 1 (●) and case 2 (Δ).

In case 1, concentrations of NH_4-N and NO_2-N and NO_3-N decreased. Conversely, concentrations of NH_4-N and NO_2-N and NO_3-N increased or remained constant in case 2. It is assumed that the metabolism of the bacteria switched from denitrification to nitrification because the condition of the experimental tank water changed to saturated dissolved oxygen concentration (Fig. 4). Therefore, we assume that the nitrogen source shifts to $NH_4^+ \rightarrow NO_2^- \rightarrow NO_3^-$ and that this trend of NO_3-N concentration is similar to NO_2-N.

In case 1, dissolved inorganic nitrogen (DIN; NH_4-N + NO_2-N + NO_3-N) shows a 90 % decrease (Fig. 9). While T-N (total nitrogen, and inorganic and organic nitrogen) decreased and was eventually no longer detected (Fig. 10)

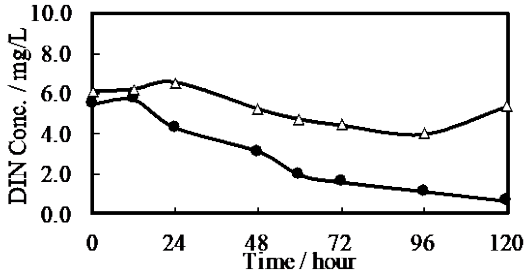

Fig. 9 Change in DIN concentration over time in case 1 (●) and case 2 (Δ).

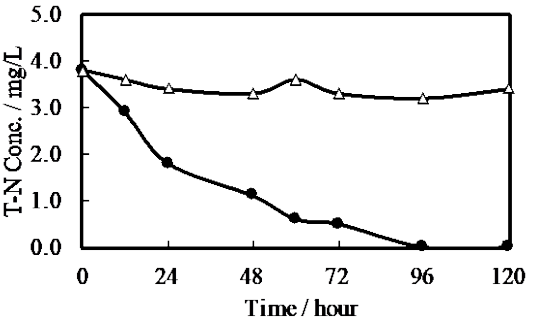

Fig. 10 Change in T-N concentration over time in case 1 (●) and case 2 (Δ).

3.1.3 Results of Bacterium concentration

DNA concentration in sludge was measured as bacterial concentration. Fig. 11 shows the change in DNA concentration. In both cases, initial concentrations of DNA are approximately 2 ug/g-sludge. In case 1, DNA doubled, while in case 2, DNA increased to 2.6 ug/g-sludge at 24 h, then decreased to 1.2 ug/g-sludge at 120 h.

The effect of the addition of *A. fecalis* to a circulation purification system on T-N concentration was investigated.

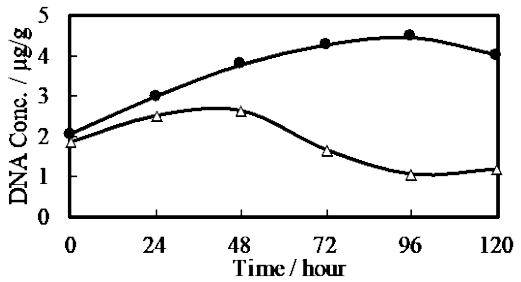

Fig. 11 Change in DNA concentration over time in case 1 (●) and case 2 (Δ).

3.1.4 Results of cesium concentration

Cesium and silica in the sludge were measured with energy dispersive X-ray analysis (EDX). The weight ratios of cesium to silica (Cs/Si) in dried sludge are shown in Fig. 12. The cesium decontamination ratio was calculated from the ratios of cesium content at 0, 6, 24, 48, 72, and 120 h as measured by EDX, using standard values for silica after measuring the weight of the dried sludge. The decontamination ratio obtained in case 1 was about 51 %.

Cesium concentrations in the water are shown in Fig. 13. In case 1, initial concentrations of Cesium are approximately 40 mg/L. Subsequently, Cesium decreases to 8.3 mg/L at 120 h indicating that 71 % of cesium was removed.

In case 2, cesium concentrations in the dried sludge and the water did not decrease. Therefore, eluted cesium from the sludge was adsorbed onto the zeolites after the sludge was decomposed by micro-bubbles and *A. fecalis*.

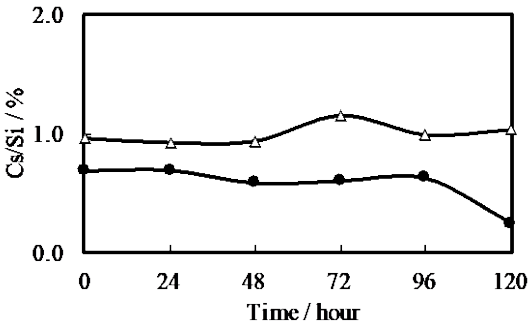

Fig. 12 Change in Cs/Si over time in case 1 (●) and case 2 (Δ) in dried sludge.

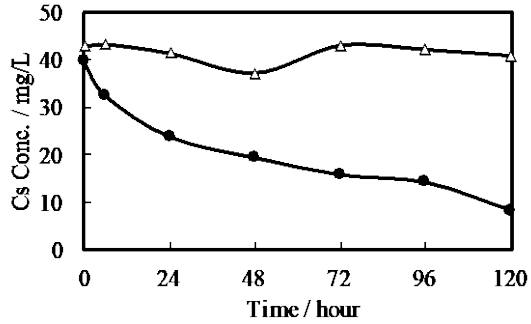

Fig. 13 Change in Cs concentration over time in case 1 (●) and case 2 (Δ) in the liquid phase.

The Model sludge was prepared 100 mg/L Cesium chloride in the tank and mixed for 24 h. So, at the time of experiment start, Cesium concentration of water was 39.6 mg/L, cesium adsorbed onto sediments was 60 mg/L. After 120 hrs, 31 mg/L (51%) of cesium onto sediments, and 8.3 mg/L of cesium in water had been remaining. Accordingly, using micro-bubble circulation system with bacteria and zeolite, 61 mg/L of cesium was removed and its removal ratio was 61%.

In case 2, using zeolite only, 42.8 mg/L of cesium in water was removed, while, cesium on

sediments remained unaltered. Therefore, total cesium removal ratio was 42.8%. Moreover, Okamoto et.al (2015) was reported that cesium concentration of water and sediments was remained unaltered using micro-bubble circulation system with bacteria [4]. These results suggest that after the deposited sludge is decomposed by our micro-bubble circulation system with bacteria and zeolite, the cesium is eluted into water, it can then be removed using zeolites.

4. CONCLUSION

We carried out elution and fixing of cesium in ocean sludge using a micro-bubble and bacterial circulation decomposition system. From water quality measurements, it can be concluded that after adding *A. fecalis*, T-N decreases and is then no longer detected. *A. fecalis* has a remarkable ability to treat samples through denitrification.

The cesium concentrations in sludge and water decreased through treatment with micro-bubbles and bacteria for 120 h. 76% of cesium in dried sludge and 51% of cesium in water were removed. From the results obtained, we can affirm that the system proposed would greatly facilitate cesium removal from ocean sludge.

5. ACKNOWLEDGEMENTS

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