Development of In-situ Radioactivity Inspection System for Radioactive Waste and Decontamination System using Micro-algae

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Abstract

In 2011, Fukushima nuclear power plant accident in Japan caused contamination of lakes and rivers due to radioactive materials released. The conventional decontamination methods using chemical precipitation and ion exchange can lower the radioactive concentration of contaminated water to a certain level. However, it is difficult to purify the large amount of contaminated water to a level lower than the very low level due to technical and economic reasons. Microalgae have recently been applied for the removal of the water-soluble radioactive materials as well as bio-energy and high-value material resource. In this study, we develop a small prototype of radioactivity measuring device using NaI(Tl) scintillator with photomultiplier tube in order to measure radioactivity in the decontamination system, which is based on semi-permeable membrane containing radionuclide-removing microalgae into radioactive contaminated water.

1. Introduction

When the Fukushima nuclear power plant accident occurred, the enormous quantities of water were used to cool the reactors. Indeed, it was estimated that approximately 250,000 tons of water were used by mid-January 2012 to cool down the nuclear reactors [1]. The general decontamination methods for the removal of radionuclides in radioactive contaminated water are mostly chemical precipitation and ion exchange, but they are costly and/or ineffective for low concentrations with significant large volume. Also, it is a time-consuming and costly process to dispose a large amount of radioactive waste generated in the dismantling of nuclear power plants. The cost of the decontamination system developed by Areva (Paris, France) and Kurion (CA,US) is around US $660 million, and 2,000 cubic meters of the generated radioactive sludge should be preserved in new long-term storages by 10 months after the accident.

Thus, the development of an effective and economical method for removing the nuclide from widespread contamination in aquatic ecosystems has become an important issue. Biosorption and phytoremediation using various biomasses have been considered as alternatives to the toxic metal removal process using chemical reagents [2-4]. Biological remediation technologies become an interesting alternative to reduce radioactive releases in aqueous effluents or to clean-up contaminated water. Lee et al. [4] reported that microalgae *Chlorella vulgaris* removes effectively radioactive cesium and strontium in contaminated water. They also discovered that biosorption of strontium induces $SrCO_3$ by photosynthetic biomimeralization.

In this paper, we introduce the outline of the development of the radioactivity decontamination system and inspection system using and the NaI(Tl) detector for the performance test and semi-permeable membrane containing radionuclide-removing microalgae into radioactive contaminated water.

2. System Design

2-1 Decontamination procedures

Figure 1 shows the principle of removal of radioactive nuclides by injecting semi-permeable membrane containing radionuclide-removing microalgae into radioactive contaminated water.

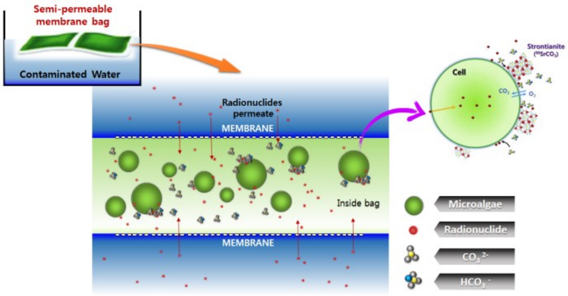


Fig. 1. Principle of removal of radionuclides using the microalgae and semi-permeable membrane

In this study, firstly, microalgae with adsorption capacity for a specific radionuclide are selected. Previous studies have shown that various microalgae have biosorption property for radioactive cesium and strontium in water. Among various microalgae, the optimal microalgae are selected through the adsorption performance tests for the radioactive species Cs-137, Sr-90 and the like. Figure 2 shows a cross-sectioned HRTEM image of *C. vulgaris* cells with strontium and conceptual model for a microalgal absorption [4].

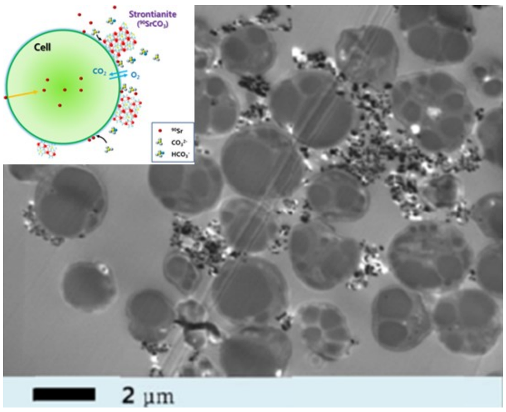


Fig. 2. A cross-sectioned HRTEM image of C. vulgaris cells and conceptual model for a microalgal absorption and surface- carbonating process [4].

Secondly, an optimal culture method capable of mass production of the selected microalgae efficiently is selected. There are two types of culture methods: Open and closed type. A culture system is constructed by selecting a culture method that minimizes the energy and nutrients to be used for microalgae cultivation while maximizing the use of CO_2.

Thirdly, a decontamination method with optimum decontamination performance is selected by applying a certain amount of cultured microalgae to radioactive contaminated water. The decontamination performance, the microbial collection rate, and the maintenance period of microalgae are compared and evaluated for various candidate methods. These methods are decontamination by adding a decontamination container to polluted water environment, by passing contaminated water through decontamination vessel, and etc.

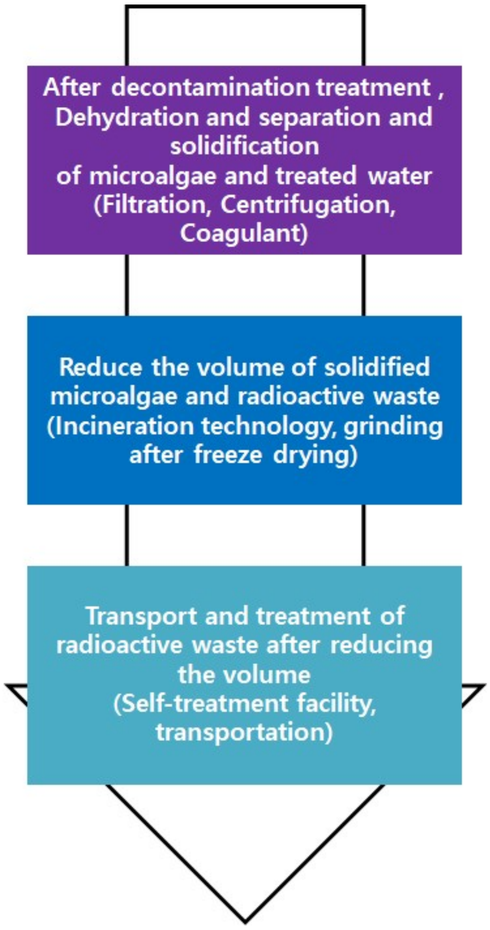


Fig. 3. A simple diagram of the post-treatment system.

Fourthly, after the decontamination is completed, the post-treatment method is selected for the bulkiness of the used microalgae. The used microalgae adsorbing radionuclides in radioactive contaminated water can be regarded as secondary radioactive waste. The contaminated microalgae, which have retained moisture, are reduced in volume and weight through post-treatment.

Figure 3 shows a simple diagram of the post-treatment system. The optimal post-treatment method is selected in consideration of the volume reduction rate, incidental waste generation rate, post-treatment time, cost and etc.

2-2. Selection of microalgae

In the previous paper [4], *C. vulgaris* cultivated in MBM and YPG media, respectively, were collected by centrifugation and washed three times with $NaHCO_3$ 3mM. The viability of microalgae interacted with intense radioactive Sr-90 and Cs-137 was evaluated in different levels of radioactive solutions for 6 days. As a result, their survival patterns were different from each other according to the radiation effect on the organisms. With high radio-resistance, *C. vulgaris* has doubled its biomass within 2 days in a malnutrition condition. When cultured under illumination, uptake of radio-strontium and radio-cesium by the microalgae was observed with time after the addition of $^{90}SrCl_2$ and $^{137}CsCl$, and maximum removals of Sr-90 and Cs-137 by C. vulgaris were ≥90% and 70% of the total Sr-90 and Cs-137, respectively. Both the microalgal cultures containing *C. vulgaris* demonstrated different efficiency for strontium uptake. These results indicate that the removal rates of strontium highly depend on microalgae strains. The removal of radio-strontium by C. vulgaris was very fast in 24h, in which almost 80% of the aqueous Sr-90 was removed from the solution. Even in 2,000Bq/ml radiation, the dissolved 90Sr decreased quickly and the Sr-90 radioactivity was little detected in solution after several days.

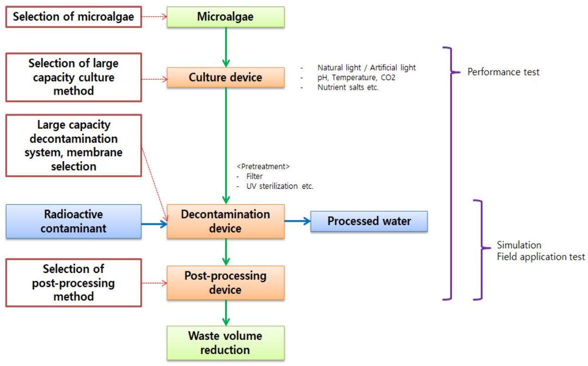
Living *C. vulgaris* had a potential to remove ^{90}Sr to very low concentrations in highly radioactive conditions. The strontium trapped by C. vulgaris was confirmed as a carbonate solid phase, strontianite ($SrCO_3$). The ^{90}Sr bio-crystallization process that becomes structurally denser by microalgae is very important to effectively grasp soluble strontium ions and to prevent ^{90}Sr resolubilization later on [4].

2-3. Conceptual design of system

The whole batch system for treating radioactive contaminated water was conceptually constructed by organically linking a microalgae mass culture system, a decontamination system, and a post-treatment system to be derived through the above-mentioned four-step study. Figure 4 shows a conceptual diagram of a system for treating radioactive contaminated water using microalgae, following the steps.

Firstly, the microalgae mass culture system produces sufficient amounts of microalgae prior to the use in the decontamination equipment. Secondly, the microalgae mass obtained through the culture system is applied to the decontamination system so that the decontamination activity can be performed at the actual site of radioactive contamination. Thirdly, before the radioactive contaminated water enters the decontamination equipment, the specific radioactivity in the water is measured and it is determined whether or not the contamination is necessary. Finally, the treated water after passing through the decontamination system is measured for specific radioactivity before releasing it outside to determine whether it is to be sufficiently decontaminated.

At this time, after the comparative tests of various detectors, a detector to be used for radioactivity measurement will be selected and applied to the system.



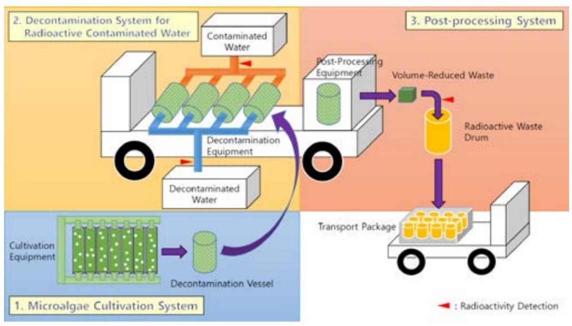


Fig. 4. Concept of radioactive contaminated water treatment system using microalgae.

2-4. Development of radioactivity detection device for decontamination system using microalgae

For the radioactivity measurement before and after the decontamination using microalgae, we developed a prototype of a small liquid radioactivity (for gamma nuclide) measurement equipment. The photograph of the prototype is shown in Figure 5. It has the capacity of 1000 ml and we plan to develop a large-scale liquid radioactivity measuring device in the future.

The developed prototype is composed of radiation detecting, shielding and analyzing parts. Lead blocks of 60mm were used to isolate from the effects of external environmental radiation or artificial radiation. A NaI(Tl) detector was used because the scintillation light yield is high and the response to electrons and gamma rays is almost linear. Also, the nuclide analyzer: 1024 channel MCA integrated with NaI(Tl) detector was used.

Fig. 5. Prototype of small liquid radioactivity (for gamma nuclide) measurement equipment using the 3inch NaI(Tl) detector and 1,000ml marinelli beaker

Figure 6 shows the linear response of sensor with gamma radioactivity using ^{137}Cs (0.662 MeV: 100, 500, 1000Bq). The radioactivity increases with increasing the linear velocity of the radiation, and thus the dose increases linearly.

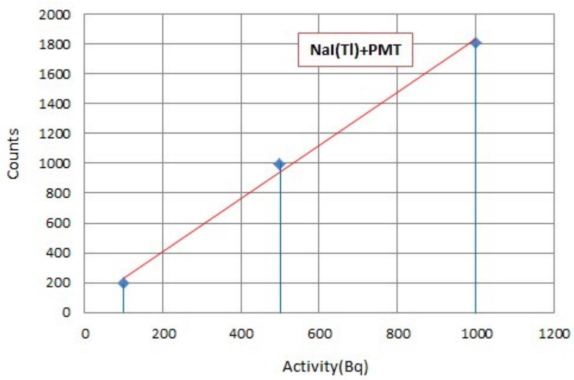


Fig. 6. Test results using the NaI(Tl) detector (0.662MeV Cs-137 : 100, 500, 1000Bq)

In the tests, the ability to measure 1/10 of the radioactivity limit recommended by the World Health Organization (WHO) for water was verified by observing the measurement of radioactivity from 1 to 1000 units. The resolution of NaI(Tl) detector was calculated to be about 8% at 0.662MeV of Cs-137.

3. Conclusion

This paper develops a small prototype of radioactivity measuring device using NaI(Tl) scintillator with photomultiplier tube for treating radioactive contaminated water using microalgae in semi-permeable membranes. This microalgae-based bioremediation with small radioactivity measuring device covers large-scale extremely low level (or low level) radioactive contaminated water.

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