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A concept of countermeasure against radioactive wastewater generated in disastrous nuclear accident such as Fukushima Daiichi site case

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1. Introduction

After the severe nuclear accident at Fukushima Daiichi nuclear power plants caused by tsunami and earthquake in March 2011, seawater was injected into the reactors for cooling, resulting in the rapid generation of a massive volume of radioactive wastewater. Before the operation of initial wastewater treatment systems supplied by AREVA and Kurion companies, which were installed about 6 months after the accident, the contaminated water was accumulated in reactor and turbine buildings, then was moved and stored in many hurriedly-prepared storage tanks including even mega float barge. The wastewater treatment systems using Cs-adsorption columns and desalination equipment was not properly operated and there were several small and big leakages of contaminated water from the wastewater treatment system and storage tanks, so that tremendous wastewater had been accumulated during those periods. That thereafter led to many secondary problems in management and treatment of the wastewater [1-3].

Since the disastrous accident at Fukushima, several measures to more enhance safety of nuclear power plants located on coastal area have been asked. As one of them, a countermeasure against generation of tremendous radioactive wastewater in disastrous nuclear accident like the Fukushima Daiichi station was asked to be prepared. This work studied a concept of countermeasure against radioactive wastewater generated in disastrous nuclear accident like Fukushima Daiichi.

2. Introduction of emergency wastewater treatment system

One of the reasons which made the wastewater treatment problem at Fukushima Daiichi site very difficult is considered to be ascribed to adaptation of storing generated wastewater in the initial stage of accident without its immediate treatment and recycling. With monitoring the progress of accident disentanglement at Fukushima and reviewing literature on treatment of radioactive wastewaters, we developed a concept to countermeasure such a case, as shown in Fig.1. The concept is to fundamentally minimize storage of wastewater generated from cooling the accident reactors by using an emergency treatment system which should be able to be rapidly installed or ready already, and effectively operated, then recycle the treated water into the reactors, even though it cannot remove completely all the radionuclides in wastewater. If an emergency system to recycle the wastewater as cooling water for crippled reactor by a quickly responsible and effective treatment way is operated from the initial stage until a normal treatment facility of contaminated water is set up, it could mitigate many secondary problems such as installation and management of many storage tanks in a wide area with potent possibility of wastewater leakage from storage facilities into environment, radiation exposure to workers to disentangle the accident, and reduce the capacity of normal wastewater treatment facility to thereafter be installed.

As the initial stage of emergency elapsing, a normal wastewater treatment facility like one installed at Fukushima Daiichi plant using many columns of adsorption and ion exchange, and several chemical unit operations is considered to be eventually required for long-term treatment of wastewater recycled and accumulated within the crippled turbine and reactor buildings. Then, the emergency wastewater treatment system can be used as a backup system against cases of trouble or leakage in the normal wastewater treatment facility.

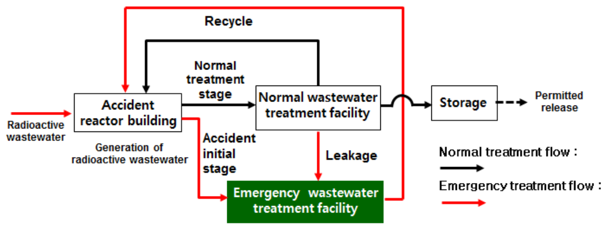

Fig.1. Concept of countermeasure against generation of huge radioactive liquid waste in disastrous nuclear accident.

3. Emergency treatment of radioactive wastewater

3.1 Major radionuclides generated in emergency case

It is necessary to know major elements in the radioactive wastewater generated in the worst nuclear accident case, their concentrations or activities. The several materials on nuclide analysis results at the wastewater treatment facility at Fukushima Daiichi reported by TEPCO (Tokyo Electric Power Company) are quite important and useful to determine the major elements. The major nuclides monitored by TEPCO in the wastewater cleanup facility to treat the contaminated water from turbine and reactor buildings of crippled Fukushima Daiichi nuclear power units were Cs-134, Cs-137, I-131, Mn-54, Co-60, Sb-125, Ru-106, etc [4,5]. They were the nuclides with high specific activities or long half-lives of over approximately one year. Unexpectedly, Sr was not monitored in the wastewater cleanup treatment facility. This may be because the Sr and Ba dissolved from corium of lava-like molten fuel contained material are co-precipitated as an isomorphous substance in seawater with sulfate anions. The TEPCO did not monitored TRU elements in the wastewater treatment system either. The reason is speculated to because the solubilities of TRU elements in the form of metal oxides are very low in the cooling water around pH 7 to 8. Based on the all the released materials, in this study, the target elements to be preferentially removed in the radioactive wastewater generated in a disastrous nuclear accident, and their concentrations were determined, as shown in Tables 1.

Table 1. Major radionuclides to be removed in wastewater treatment system in case of disastrous nuclear accident and their estimated concentrations

[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
[illegible]	2.1	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]
[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]	[illegible]

3.2 Chemical unit operation for emergency wastewater treatment system

The way to treat the radioactive wastewater massively-generated in emergency case in Fig.1 has to have several characteristics such as easy operation, quick response, and high decontamination yield in its application. Based on survey of literature on wastewater treatment, precipitation can meet such conditions relatively well compared with other chemical unit operations such as column operation using adsorbents or ion exchangers, evaporation, reverse osmoses, ultrafiltration, etc. [6-8]. Cs, Sr, and I ions have been generally treated by adsorption in column. Zeolite and metal ferrocyanide for Cs, zeolite and barium sulfate for Sr, and active carbon and active alumina for I are used as their adsorbents. The system consisting of columns with such adsorbents is considered to be not suitable in emergency case in views of its preparation time and complicate operation. Therefore, in this study, adsorption-precipitation way for removal of such elements was selected so that suitable adsorbents for each element are directly used in the form of powder itself in solution instead of using them in columns. The other radioactive substances of Co, Mn, Ru, Sb, and suspended fine particles remained in solution after using the adsorbents can be removed together from the solution by conventional coagulation-flocculation using ferric or aluminum hydroxide and organic flocculants. Cs, Sr, and I, however, cannot be properly removed by such as coagulation-flocculation. When precipitation method is applied to treat effectively and rapidly such a radioactive wastewater, it has to have the characteristics of high decontamination yield and rapid settlement of absorbents in particle and formed flocs. Based on above-mentioned concept, a sequential precipitation process for emergency radioactive wastewater treatment system in Fig.1 was suggested, as shown in Fig.2. In the process, oil and other suspended materials should be first to be separated by pressure floatation, strainer, etc., then the sequential precipitations consisting of adsorptions-precipitation for Cs, S,r, I, and flocculation-precipitation for the residual elements are carried out in the a few sets of combined mixing and settling vessels for each precipitation step, and finally the supernatant coming out of the precipitation stage is desirable to be treated by micro-filtration or ultra-filtration to remove suspended substances of fine flocs or colloidal particles remained in solution.

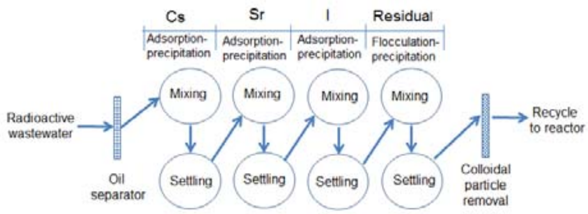

Fig.2. Sequential precipitation process for emergency treatment of huge and radioactive wastewater.

In this work, the adsorption precipitations for removals of Cs, Sr, and I, and the flocculation-precipitation for removal of residual elements of Co, Mn, Sb, Ru were experimentally studied in views of their decontamination yields and settling speeds to evaluate whether the concept and method suggested in Fig.1 and Fig.2 for emergency treatment of radioactive wastewater could be embodied.

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