REMOVAL OF CO^{60} FROM THE LIQUID RADIOACTIVE WASTE IN HIGH SALT CONCENTRATION

Sung Paal Yim, Young Min Kim, Hun Hwee Park, Jae In Shin and Joon-Hyung Kim
Korea Atomic Energy Research Institute
P.O.Box 7, Daeduk-Danji
Taejon, 305-353, Korea(R.O.K.)

ABSTRACT

The removal of Co^{60} from the liquid radioactive waste in high salt concentration was studied. The removal efficiency of precipitation for the radionuclides and the filtration property of precipitate were investigated over the waste in the high salt concentration caused by the seawater.

From the experimental results it can be concluded that Co^{60} in the liquid radioactive waste in high salt concentration is effectively removed by precipitation resulting from the addition of alkali and filtration of precipitate. The poor filtration property of precipitate was improved by the use of appropriate polymer flocculant.

INTRODUCTION

The accident can occur that the seawater being used as coolant at nuclear power plants is contaminated by leakage of the radioactive materials or, inversely, the seawater is flooded into a system handling the radioactive materials, although rarely. If it happens, a large volume of the liquid radioactive waste in high salt concentration is generated. The waste is not effectively decontaminated with the usual treatment method such as evaporation and ion-exchange, which show poor decontamination, low volume reduction and various operational problems because of high salt concentration of the waste. It is known that several commercialized selective ion-exchangers can effectively decontaminate the liquid radioactive waste in high salt concentration (1,2). However, the selective ion-exchangers are not economical since they presently cost high, and they were also not effective on the removal of some specific radionuclide such as cobalt.

It has been studied in the field of water treatment that some component of the seawater remove the contaminant from the water or the waste solution by co-precipitating under the basic condition (3,4). Especially, the negatively charged colloids and several anions are well adsorbed on the positively charged surface of $Mg(OH)_2$ particles produced by precipitation of the seawater (5). However, it is not known whether or not the radionuclides, existing mostly in the form of cation, can be removed with precipitate produced from the seawater. If this precipitate incorporate the radionuclides, the liquid radioactive waste in high salt concentration (the seawater contaminated with the radionuclides and the liquid radioactive waste mixed with the seawater) can be easily decontaminated by simply addition of alkali. In this study, it was experimented that the liquid radioactive waste in high salt concentration may be effectively decontaminated by precipitation due to addition of alkali and filtration of resultant precipitate.

EXPERIMENTS

Materials and Reagents

The simulated liquid radioactive waste was used in this experiment. This solution is prepared by addition of small volume of radioactive solution containing Co^{60}(II), Sr^{85}(II) and Cs^{137} ions into the seawater or the seawater diluted with the demineralized water. The specific radioactivities of Co^{60}, Sr^{85} and Cs^{137} are 3.7×10^3 Bq/mL, respectively. The seawater was collected at Ah-San bay in Korea, on June 1992 and its composition is presented in Table I. Reagents used are all first-graded ones. The polymer flocculant is Yangfloc A-701 (anionic polyelectrolyte) made by Eyang Chem. Co. in Korea. Co^{60}, Sr^{85} and Cs^{137} contents in the solution were determined by a multichannel analyzer Canberra Series 35 plus with NaI detector.

TABLE I
Composition of Seawater Used

Ion	Concentration(ppm)
Na^+	10,040
Mg^{2+}	1,210
Ca^{2+}	380
K^+	360
Cl^-	18,050
SO_4^{2-}	2,500

Experimental Methods

Precipitation is induced by rapid addition of a known volume of alkaline solution into the liquid radioactive waste. Suspension is stirred with jar tester (rotation speed = 300 rpm). The suspension is centrifuged (g = 5,000, time = 10 min), and the supernatant is analyzed on a gamma spectral analyzer.

Filtration test is carried out in order to compare the filtration properties of suspensions before and after the addition of polymer flocculant. Flocculation is performed by adding a pre-determined volume of polymer flocculant solution into the suspension. The suspension is stirred intensively (500 rpm) during the first 10 seconds to homogenize the solution and then slowly during the next 10 minutes to prevent breakage of floccules. The filtration properties of suspensions are observed with Filtration-Permeatry (6).

RESULTS AND DISCUSSIONS

Precipitation

Cations and anions are much included in the seawater as presented in Table I. The amount of cations included in the seawater decreases in order of Na^+, Mg^{2+}, Ca^{2+}, etc. It is already well known that Mg^{2+} and Ca^{2+} ions in the seawater are precipitated in the form of $Mg(OH)_2$ and $CaCO_3$ in the basic condition (7). The pH change of the seawater according

to the addition of sodium hydroxide is presented in Fig. 1. The precipitate is formed at the range of pH 10.4 - 11. In Fig. 2, the changes in the amount of Mg^{2+} and Ca^{2+} ions are presented when NaOH is added to the seawater. Fig. 3 shows the results of X-ray diffraction analysis for precipitate produced. It is known from the results that most of the precipitate are $Mg(OH)_2$.

Effect of Precipitation on Decontamination

In Fig. 4 are presented how much of Co^{60} are removed when alkali are added to the sea water contaminated with the radionuclide. The removal efficiency rapidly increases until the addition of 3 meq NaOH/L, and slowly increases after the addition of 5 meq NaOH/L. The removal efficiency for Co^{60} is about 90% at the addition of 2 meq NaOH/L, and reaches to about 99.5%, 3 meq NaOH/L. The maximum removal efficiency for Co^{60} is more than 99.8 % and this means that decontamination factor of more than 500 can be obtained by this method. Experiments with another kind of alkali such as CaO and $Ca(OH)_2$ also show the same results with sodium hydroxide. The removal efficiency for Co^{60} is, in Fig. 5, presented when the concentration of the sea water in the radioactive solution is changed. The conditions of precipitation are constantly maintained at pH 10.8. The result shows that Co^{60} can be removed more than 99 % by precipitation even when the concentration of the sea water in the solution is 5 %.

It is very interesting that Co^{60}, existing mostly in the form of cation, can be exceedingly incorporated and removed with the positively charged $Mg(OH)_2$ particles produced by precipitation of the seawater.

However, the other kinds of radionuclides, Cs^{137} and Sr^{85}, are not removed with precipitate at all.

In addition, the effect of boric acid, one of the major component of radioactive waste at PWR plants, on the removal of Co^{60} was investigated. Boric acid does not directly affect the removal of Co^{60} in the radioactive solution but more alkali is required in order that precipitation is occurred because alkali is exhausted by reaction with boric acid.

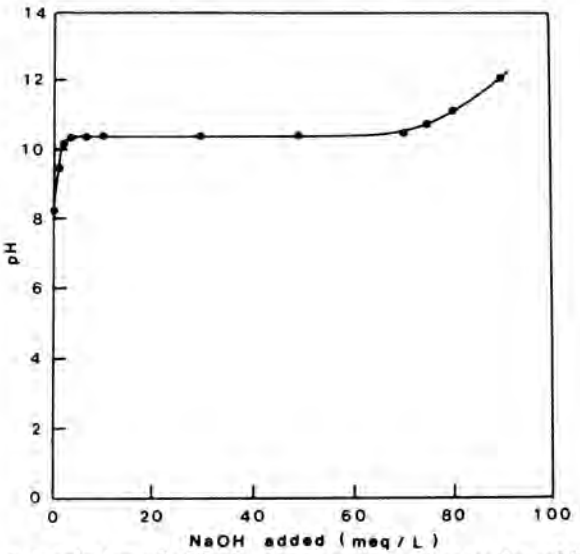

Fig. 1. Variation of pH of seawater according to the addition of NaOH.

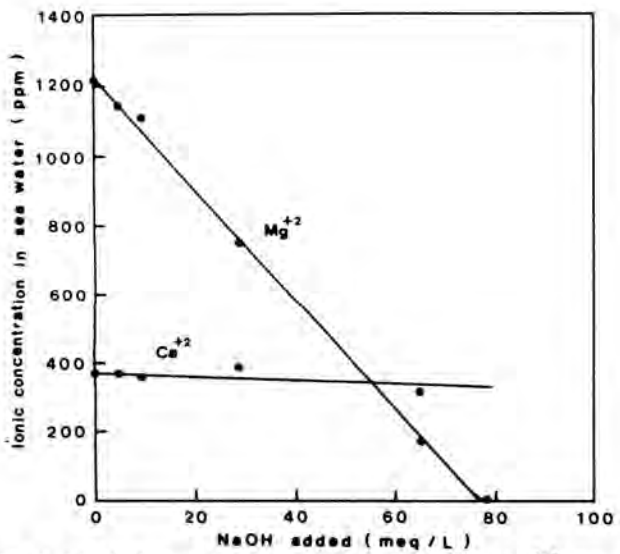

Fig. 2. Variation of ionic concentration for Mg^{+2} ion and Ca^{+2} ion in the seawater according to the addition of NaOH>.

Filtration of Precipitate

The precipitate should not be released with decontaminated solution since the radioactivity of the precipitate is very

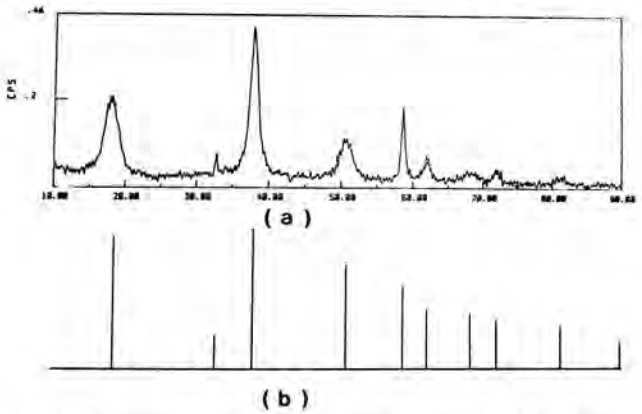

Fig. 3. XRD patterns of (a) the precipitate and (b) the standard $Mg(OH)_2$.

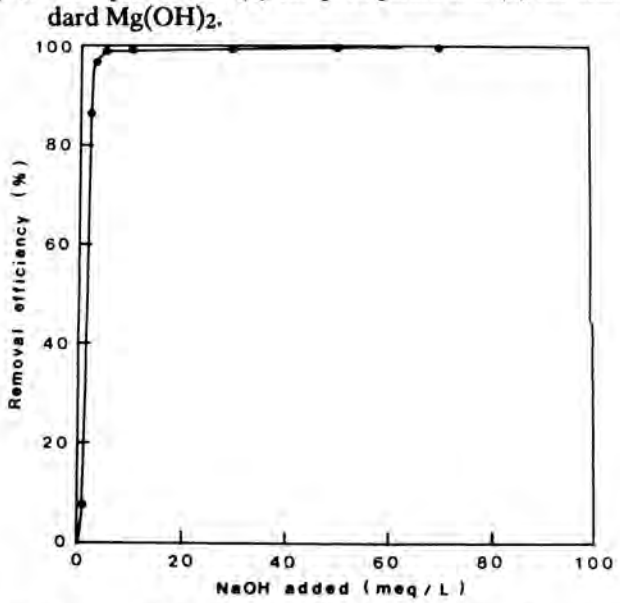

Fig. 4. Removal efficiency of Co^{60} by precipitation of seawater.

high. However, since the precipitates, $Mg(OH)_2$ and $CaCO_3$, are taken long time to be settled and difficult to be filtrated because of their high tendency of dispersion and high specific resistance of filtration. These problems can be improved by the use of appropriate polymer flocculant to aid agglomeration of suspended particle.

The results of filtration tests are presented in Fig. 6. The value of the specific resistance of filtration of precipitate is 1.6×10^{14} m/Kg. This means that precipitate is very difficult to be filtrated. Agglomerate of precipitate is formed when anionic polymer flocculant is added, and its specific resistance of filtration is around 10^{12} m/Kg. The results indicate that filtration property of suspension is improved more than 100 times by adding the flocculant. The effect of anionic polymer coagulant appears very effectively when the amount of anionic polymer coagulant are added in the range of 30 - 60 ppm.

CONCLUSIONS

As a result of experiment, it can be concluded that Co^{60} in the seawater contaminated with the radionuclides and the liquid radioactive waste mixed with the seawater are effectively removed by precipitation resulting from the addition of alkali and filtration of precipitate. The poor filtration property of precipitate is improved by the use of appropriate polymer flocculant to aid agglomeration of suspended particle. However, the other radionuclides cannot be removed in this experiment. Nevertheless, this method will be applied as a process to decontaminate the radioactive waste in high salt concentration in case that the removal of radioisotopes of cobalt become a serious problem.

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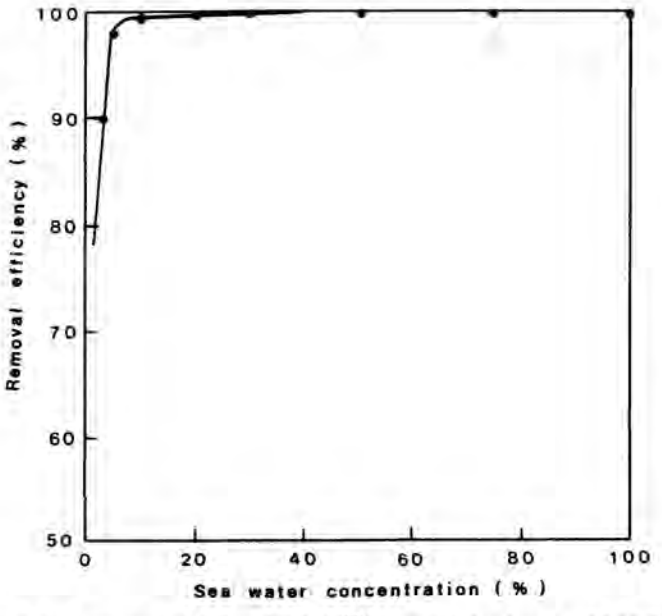

Fig. 5. Removal efficiency of Co^{60} by precipitation at pH 10.8 according to the seawater concentration of the solutions.

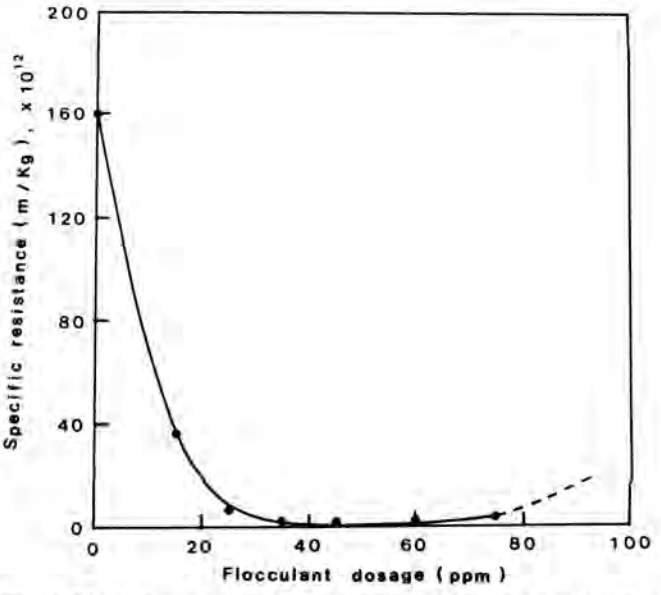

Fig. 6. Variation of specific resistance of filtration according to the polymer flocculant dosage.