

## Removal of cesium from the radioactive waste solution by sorbent or precipitant

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### 1. Introduction

The breakage of a storage tank and connection tubes in the underground nuclear industry facilities would contaminate the surrounding land with radioactive nuclides. When acid or alkaline solution is used for the remediation of a contaminated soil, radioactive waste solution will be produced, and it contains various kinds of metal ions as well as radioactive nuclides.

In order to remove radioactive nuclides from the waste solution, several techniques such as precipitation, sorption, ion exchange, and evaporation/concentration of radioactive nuclides have been used. To select more effective one from these treatment methods, a proper reagent and the optimal condition should be obtained. In this study, sodium tetraphenylborate (Na-TPB), zeolite (mordenite), nickel hexacyanoferrate and Prussian blue as a precipitant or sorbent for radioactive cesium were added into the Cs-contaminated solution, respectively, and their results are compared one another. And their optimal experimental conditions were also studied.

### 2. Experimental and Results

#### 2.1. Comparison with the removal efficiencies of cesium by precipitant and sorbent

Cs-contaminated soil was put into the water, and pH of the solution was adjusted to 0 by adding sulfuric acid. After a 3-hours shaking, the solution was centrifuged for 20 minutes at 1300 rpm. The supernatant was collected, and it was used as a Cs-contaminated waste solution for this study.

As a precipitation agent or sorbent for cesium, 50 mg of Na-TPB, zeolite, nickel hexacyanoferrate and Prussian blue were added into 100 mL of waste solution, respectively. The pH of solution was adjusted by adding calcium oxide. The solution was stirred for two hours at 300 rpm using a magnetic stirrer. Then, the solution was passed through a Whatman filter paper No. 4 (pore size 25  $\mu$ m). Finally, the activity of cesium in eluent was analyzed by using a  $\gamma$ -spectrometry.

The experimental results for a cesium removal by precipitants and a sorbent are shown in Table 1. This table indicates that the cesium concentration is changed in the order of nickel hexacyanoferrate > Na-TPB > Prussian blue > Zeolite

Table 1 The comparison of the removal efficiency of cesium by precipitant or sorbent.

Precipitant or sorbent	Initial cesium conc. (Bq/mL)	pH	Added amount (mg)	Cesium conc. after treatment (Bq/mL)
Na-TPB	4.4	11	50	0.87
Zeolite	3.2	7	50	1.8
Nickel hexacyanoferrate	4.4	7	50	0.036
Prussian blue	6.0	7	50	2.4

#### 2.2. The variation of cesium concentration upon amount of precipitants

The variation of cesium concentration was examined with the amount of nickel hexacyanoferrate which has the highest removal efficiency of cesium among the precipitants and a sorbent used in this work. 5~100 mg of nickel hexacyanoferrate was added into 100 mL waste solutions, and the solutions were adjusted pH=7. When 5 mg of nickel hexacyanoferrate was added into the waste solution with 4.4 Bq/mL of cesium, the concentration of cesium in the supernatant decreased to 0.22 Bq/mL as shown Table 2. This concentration is higher than our self-disposal limiting value for cesium, 0.1 Bq/mL. However, the addition of 30 mg of nickel hexacyanoferrate decreased the concentration of cesium in the supernatant to 0.040 Bq/mL, which corresponds to about 99% of cesium removal from the waste solution. Though the addition of 50 and 100 mg of nickel hexacyanoferrate reduced the cesium concentration to 0.036 and 0.031 Bq/mL, respectively, the difference of cesium concentration was insignificant in comparison with the addition of 30 mg of it. Therefore, 30 mg of nickel hexacyanoferrate is considered as a proper amount to remove cesium from the 100 mL of waste solution with less than 4 Bq/mL of cesium. However, when initial concentration of cesium was 8.5 Bq/mL, 50 mg of nickel hexacyanoferrate was required to reduce cesium concentration to below 0.1 Bq/mL.

For comparison with nickel hexacyanoferrate, Na-TPB was also added into a Cs-contaminated solution. Generally Na-TPB is known to react with cesium very well at around pH 11. So, the pH of solution was

adjusted to pH 11 with CaO. When 30 ~100 mg of Na-TPB was added into 100 mL of waste solutions with 4.4 and 8.5 Bq/mL of cesium, all cesium concentration in the supernatant after precipitation was greater than 0.1 Bq/mL (Table 2). This result also indicates that nickel hexacyanoferrate is the more efficient than Na-TPB for the removal of cesium from the Cs-contaminated water.

Table 2 The removal of cesium by Nickel hexacyanoferrate and Na-TPB.

Precipitants	Initial cesium conc. (Bq/mL)	pH	Added amount (mg)	Cesium conc. after treatment (Bq/mL)
Nickel hexacyanoferrate	4.4	7	5	0.22
			30	0.04
			50	0.036
			100	0.031
	8.5		30	1.2
			50	0.025
Na-TPB	4.4	11	30	1.3
			50	0.87
			100	0.16
	8.5		30	8.4
			50	7.1

### 2.3. pH effect

To get an optimum pH condition for the removal of cesium from a waste solution by nickel hexacyanoferrate, the precipitation treatment was performed at pH of 5.5, 7.0 and 8.5. CaO and 0.1M HNO<sub>3</sub> were used to adjust pH. Table 3 was obtained from the experiment, and it shows the greatest decrease of cesium concentration at pH 7.

Table 3 The change of cesium concentration by nickel hexacyanoferrate at various pH conditions.

pH	Initial cesium Conc. (Bq/mL)	Added amount (mg)	Cesium conc. after treatment (Bq/ml)
5.5	4.4	30	0.35
7	4.4	30	0.01
8.5	4.4	30	0.07

### 3. Conclusions

The removal efficiencies of cesium from a waste solution by Na-TPB, zeolite, nickel hexacyanoferrate, Prussian blue were compared. Among these reagents, nickel hexacyanoferrate was selected the best precipitant, and optimal removal was occurred at pH 7. On the basis of these results, further researches such as the characterization of a precipitate, the effect of a particle size and a reaction temperature, the hindrance

of competitive ions, etc. would be performed in future in order to get a more optimal removal condition.

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