

## **Soil Washing Experiment for Decontamination of Contaminated NPP Soil**

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### **ABSTRACT**

The preliminary experiment was performed to obtain the operating conditions of soil washing decontamination process such as decontamination agent, decontamination temperature, decontamination time and ratio of soil and decontamination agent. To estimate decontamination efficiency, particle size of soil was classified into three categories;  $\geq 2.0$  mm,  $2.0 \sim 0.21$  mm and  $\leq 0.21$  mm. Major target of this experiment was decontamination of Cs-137. The difference of decontamination efficiency using water and neutral salts as decontamination agent is not high. It is concluded that the best temperature of decontamination agent is normal temperature and the best decontamination time was about 60 minutes. And the best ratio of soil and decontamination agent is 1:10. In case of Cs decontamination for fine soils, the decontamination results using neutral salts such as  $\text{Na}_2\text{CO}_3$  and  $\text{Na}_3\text{PO}_4$  shows some limits while using strong acid such as sulfuric acid or hydrochloric acid shows high decontamination efficiency ( $\geq 90\%$ ). But we conclude that decontamination using strong acid is also inappropriate because of the insufficiency of decontamination efficiency for highly radioactive fine soils and the difficulty for treatment of secondary liquid waste. It is estimated that the best decontamination process is to use water as decontamination agent for particles which can be decontaminated to clearance level, after particle size separation.

### **INTRODUCTION**

Small amounts of radioactive particles released from NPP are deposited onto soils inside NPP and concentrated into chimneys of drainage and other soils due to the migration of nuclide by ground water or rain. And fine contaminated soil and sludge can be accumulated in NPP. Therefore the rise of contamination level in particular region can be caused after operation of NPP.

To solve these problems caused during normal operation, contaminated soils and sludge should be collected and isolated from environment. And these soils and sludge should be treated proper process and in case of under clearance level after treatment, these can be self-disposed into the NPP area. Otherwise these should be drummed and kept in Radioactive waste Building. Accumulation of contaminated soils in NPP is inevitable and an amount of these materials contaminated by released radioactive materials during operation and decommissioning are also generated in decommissioning stage. Therefore it is necessary to minimize the amount of contaminated soils and sludge through appropriate decontamination process.

Among several decontamination processes used for contaminated soil, soil washing process is selected as decontamination process coupled with particle separation.[1-7] In this study, several experiments are performed to obtain operating conditions of soil washing decontamination process such as decontamination agent, decontamination temperature, decontamination time and ratio of soil to decontamination agent.

## **MATERIALS AND METHODS**

### **Materials**

Experiment was performed using soils of which surface dose rate was  $0.1\text{mRh}^{-1}$ . These soils had been kept inside storage facility of NPP. First, these soils were separated according to the particle size and analyzed using GeLi detector. Major nuclides were Co-60, Cs-134 and Cs-137. Radioactivity was not detected in particles above 4.76 mm, coarse sand, and radioactivity was increased as particle size was decreased.

Main contaminant was Cs-137 and an amount of Cs-134 was also detected. Radioactivity of Co-60 was very small and decontamination of Co-60 was easy. In this study, Cs-137 was selected as target nuclide for decontamination because of decontamination difficulty and large amount. And soils of which particle size was below 2 mm were selected.

### **Experiments**

Soils were separated according to the particle size and 10 g of soils below 2.0 mm of particle size were added to 100 g of decontamination agents. The soils were decontaminated about 2 hours using several decontamination agents at room temperature. Shaker was used to enhance the contacts of soil and agent. After soil was sunk, agents were separated from mixture. To remove decontamination agents from soils completely, soils were cleaned with distilled water. After depressurizing filter was used to acquire silt/clay component of soils, soils were placed into the Petri-dishes and completely dried at  $40^\circ\text{C}$ . Finally, residual radioactivity in soil was analyzed and decontamination efficiencies were acquired.

To decide optimum decontamination conditions, experiments were performed with changing decontamination time, ratio of soil to agent and temperature of agent. To acquire more precise operation conditions for design of decontamination equipment, particle size of soil was classified into three categories;  $\geq 2.0$  mm,  $2.0 \sim 0.21$  mm and  $\leq 0.21$  mm. In these cases, soils were decontaminated using water at room temperature and the effects of time and ratio were investigated.

## **DISCUSSIONS**

### **Decontamination agent**

Fig. 1 shows the efficiencies of several agents for Cs-137 decontamination. Only Cs-137 decontamination efficiencies are shown because total decontamination efficiency and reduction efficiency of contaminated soils are varied according to Cs-137 decontamination efficiency which is difficult to decontaminate. Initial radioactivity of Co-60 is very low and radioactivity is

not detected after decontamination. Cs-134 shows similar tendency to Cs-134. As shown in Fig. 1, strong acids such as  $\text{H}_2\text{SO}_4$ ,  $\text{HCl}$  and  $\text{HNO}_3$  show high decontamination efficiency but  $\text{Na}_2\text{CO}_3$  is considered as good agent because of acids' toxicity.

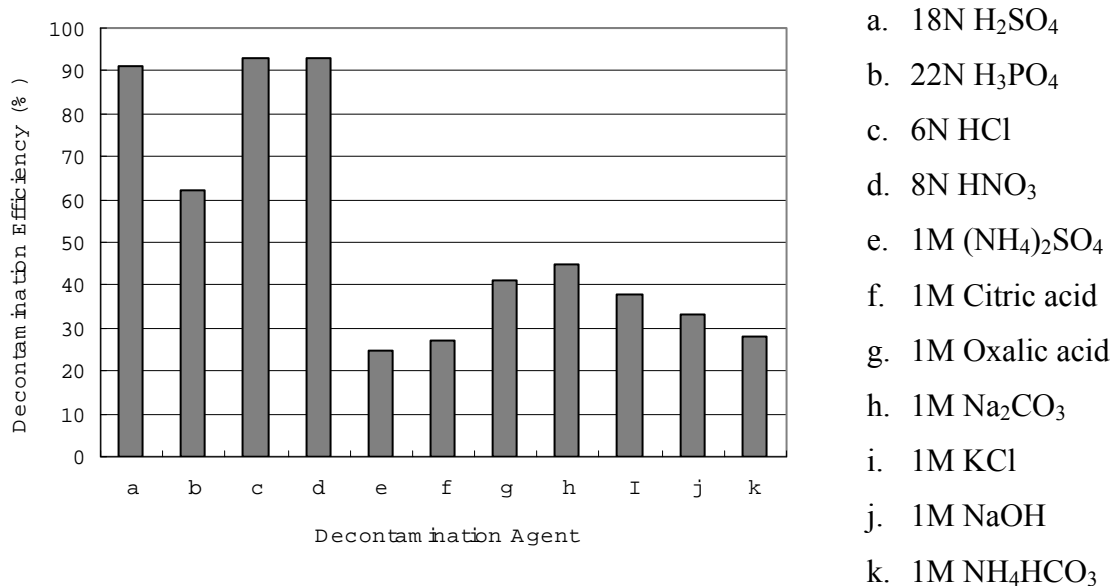


Fig. 1. Decontamination efficiency of each decontamination agents

### Decontamination Time

To investigate the decontamination efficiency according to time, decontamination times are changed to 30 min, 60min and 120min. Fig. 2 shows the decontamination efficiency of  $0.1 \text{ mR h}^{-1}$  soil according to time. The ratio of agent to soil was set to 10:1 and the efficiencies according to agents' temperature were also investigated. Results of 120 min showed about 10% higher efficiencies than results of 30 min. Selected agents were  $\text{Na}_2\text{CO}_3$ ,  $\text{Na}_3\text{PO}_4$  and water according to the result above and literature survey. Under the same condition, 1M  $\text{Na}_2\text{CO}_3$  shows the highest decontamination efficiency but the efficiency was not much higher than another agents.

These results mean that decontamination efficiencies of neutral salt such  $\text{Na}_2\text{CO}_3$  and  $\text{Na}_3\text{PO}_4$  show some limit for  $^{137}\text{Cs}$  decontamination. As shown in Fig. 1, the efficiencies of acids such as citric acid and oxalic acid were inferred as 40%. Therefore decontamination of soil using chemical agents was considered inappropriate. Of course, strong acids such as  $\text{H}_2\text{SO}_4$  and  $\text{HCl}$  show higher efficiency rather than 90%. But strong acids did not show sufficient efficiency in case of highly radioactive fine sand and generate secondary liquid waste which is difficult to treat. Therefore, decontamination using strong acid was also considered inappropriate.

### Ratio

In order to investigate efficiencies according to the ratio of soil to agent, experiments were performed to the ratios of 10:1 and 5:1, respectively. Fig. 3 shows little difference on efficiency at room temperature but shows the rising of efficiency as temperature was rising and the amount of agent was increasing.

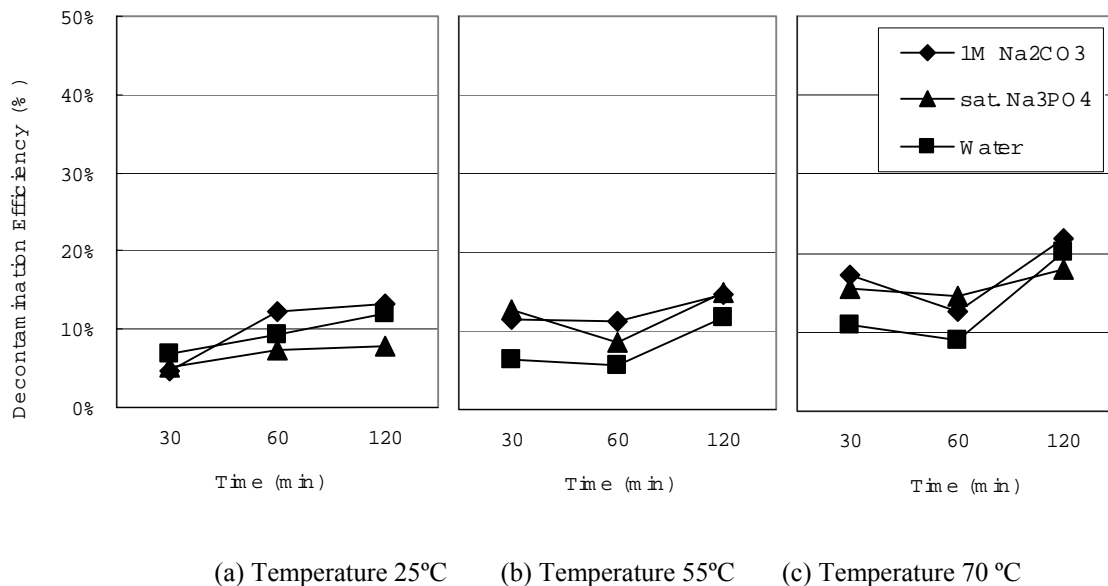


Fig. 2. Decontamination efficiency according to time ( $0.1\text{mRh}^{-1}$ , 9.1% solids)

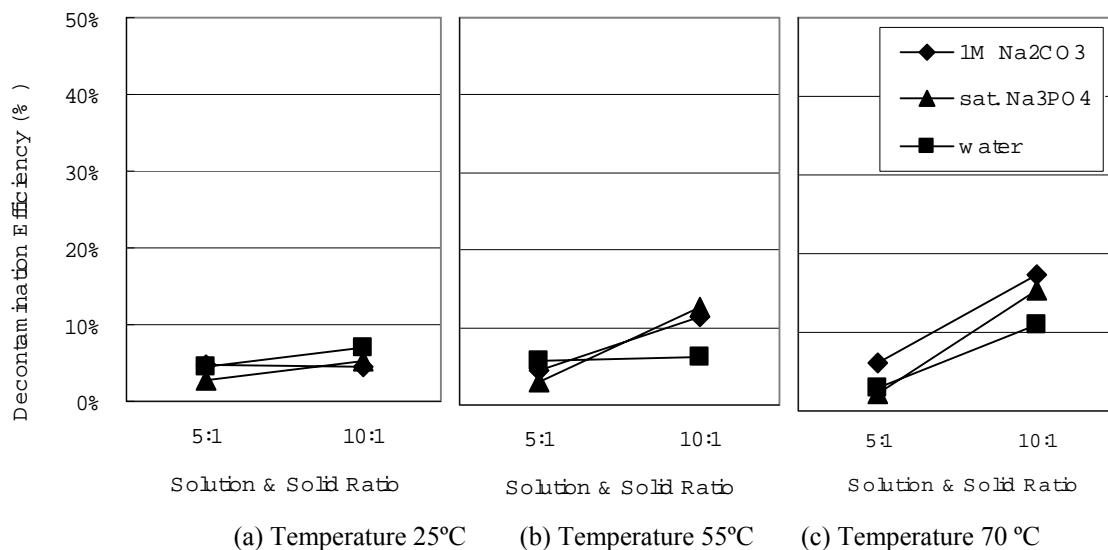


Fig. 3. Decontamination efficiency according to ratio ( $0.1\text{mRh}^{-1}$ , 30min)

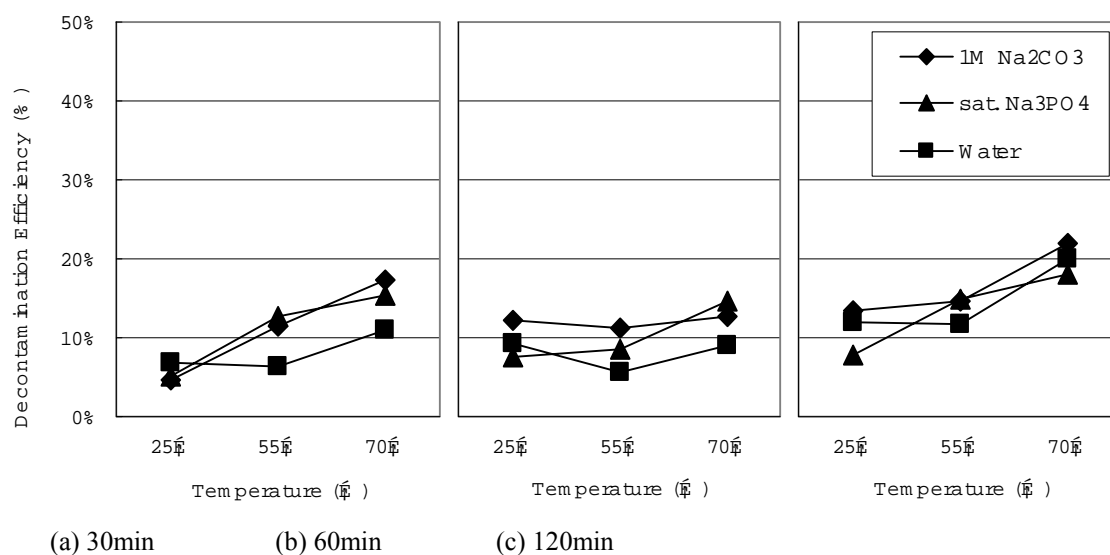


Fig. 4. Decontamination efficiency according to temperature ( $0.1\text{mRh}^{-1}$ , 9.1% solids)

### Temperature

Fig. 4 shows the decontamination efficiency according to temperature of agent. When decontamination time was 30 min and 120 min, rising of efficiency was shown. But there was little difference on efficiency at 60 min of decontamination time.

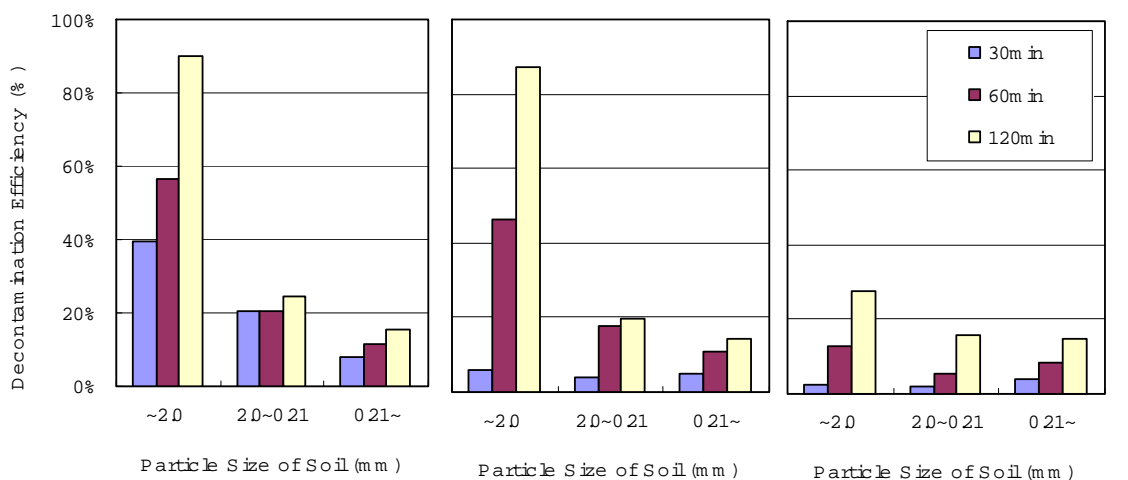
### Particle Size

To acquire more precise operation conditions for design of decontamination equipment, particle size of soil was classified into three categories;  $\geq 2.0$  mm,  $2.0 \sim 0.21$  mm and  $\leq 0.21$  mm. Under the assumption that decontamination equipment uses water as decontamination agent at room temperature, experiments were performed using water as agent at room temperature.

Decontamination times were selected 30 min., 60min. and 120 min. and ratios were selected 5:1, 10:1 and 20:1. As shown in Fig. 5(a), the efficiency of soils above 2.0 mm of particle size was about 90% using water at room temperature and decontamination time was 120 min.

Decontamination efficiencies were decreasing as particle size of soil were decreasing. The efficiencies of soils below 0.21 mm of particle size were about 15%. As shown in Fig. 5(b), efficiencies shows similar tendency to (a) except for 30 min. In Fig. 5(c), efficiencies according to the size of soil show little difference because of small agent amount.

The efficiency of soils above 2.0 mm of particle size was good using only water but the efficiency of soils below 2.0 mm of particle size was low. In case of  $0.1\text{mRh}^{-1}$  soil above 2.0 mm of particle size, it is possible to decontaminate soil to 90% if decontamination time is enough. This means that about 90% of cesium is bounded to soil with lower binding force or surface precipitation. But maximum decontamination efficiency is 70% if dose rate is increased to  $1.0\text{mRh}^{-1}$ . This means that binding force of cesium is higher as dose rate is higher.



(a) Solution & Solid Ratio 20:1 (b) Solution & Solid Ratio 10:1 (c) Solution & Solid Ratio 5:1

Fig. 5. Decontamination efficiency according to particle size of soil ( $0.1\text{mRh}^{-1}$ )

## CONCLUSION

The preliminary experiment was performed to obtain the operating conditions of soil washing decontamination process such as decontamination agent, decontamination temperature, decontamination time and ratio of soil and decontamination agent. To estimate decontamination efficiency, particle size of soil was classified into three categories;  $\geq 2.0$  mm,  $2.0 \sim 0.21$  mm and  $\leq 0.21$  mm. Major target of this experiment was decontamination of  $^{137}\text{Cs}$ .

The difference of decontamination efficiency using water and neutral salts as decontamination agent is not high. It is concluded that the best temperature of decontamination agent is normal temperature and the best decontamination time was about 60 minutes. And the best ratio of soil and decontamination agent is 1:10.

In case of Cs decontamination for fine soils, the decontamination results using neutral salts such as  $\text{Na}_2\text{CO}_3$  and  $\text{Na}_3\text{PO}_4$  shows some limits while using strong acid such as sulfuric acid or hydrochloric acid shows high decontamination efficiency ( $\geq 90\%$ ). But we conclude that decontamination using strong acid is also inappropriate because of the insufficiency of decontamination efficiency for highly radioactive fine soils and the difficulty for treatment of secondary liquid waste.

It is estimated that the best decontamination process is to use water as decontamination agent for particles which can be decontaminated to clearance level, after particle size separation.

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