Removal characterization of $^{133}$Cs and $^{127}$I in a water treatment process using a laboratory scale experiment

Hee Suk Lee, Jeongyup Lee, Byengsuk Yoon, Youjin Yim, Ilhwan Choi, Hyukjin Cho, Songhee Lee, Kyounghee Baik, Ju Hyeon Park and Yu Jeong Huh

ABSTRACT

Due to the tragic disaster that happened in Japan and crippled the Fukushima nuclear power plant, serious concerns have been raised regarding the contamination of drinking water as a result of the radioactive materials that were released. Even though the quantities of radioactive material in rain were relatively low, people were concerned about the drinking water. Therefore, there is a need to know the removal efficiency of the unit process of water treatment and to prepare a safety plan to protect the public’s health from radioactive materials. In this study, the laboratory scale removal rates were estimated for the coagulation/flocculation, adsorption, and ion exchange processes. The reference standard materials which are stable elements, Cesium-133 (Cs-133) and Iodine-127 (I-127), were used for the typical and advanced water treatment processes at the laboratory scale. For the coagulation/flocculation process, three major coagulants were assessed for this process. However, the removal rates of this process were low. For the adsorption process, powdered activated carbon and zeolites were investigated. The powdered activated carbon showed insignificant removal rates for both reference materials. However, synthetic zeolite was an effective process for Cs-133, and the ion exchange method showed high removal rates for both Cs-133 and I-127.

Key words | jar-test, reference standards, removal efficiency, water treatment

INTRODUCTION

After strong earthquakes hit Japan on March 11, 2011, a massive tsunami followed and struck the northeast coast of Japan. Following the earthquakes and tsunami, the Fukushima nuclear power plant was critically damaged, causing officials to employ an emergency shutdown of the reactors, and radioactive materials were released into the environment. Evidence of radioactive material fallout has been reported across the country and has been documented (Bolsunovsky & Dementyev 2011; Bowyer et al. 2011; Manolopoulos et al. 2011). The evidence detailed in the literature regarding radioactive material has resulted in public health precautions all over the world. Due to the public health risks posed by the threat of radioactive materials in the environment, public health policies have been significantly impacted even though there is no clear evidence of any major health effects on the public due to low levels of radioactivity. In the aftermath of the radioactive material fallout, radioactive materials have been detected in the tap water of the northeast region of Japan, as reported by the public media. The major radioactive materials found were Cesium-137 and Iodine-131. Subsequently, radioactive materials were detected in tap water and various foods, such as plants and vegetables, which in turn dramatically increased people’s concerns about radiation hazards. By the end of March 2012, low levels of radionuclide materials such as Cesium-134, Cesium-137, and Iodine-131 were detected in the ambient air and rainwater of Korea. People have been concerned about the drinking water, which has...
been treated with the conventional purification process. Even though treatment efficiencies for radioactive materials have been evaluated for several processes (Summers et al. 1988; Goossens et al. 1989; Sinha et al. 1995; Gafvert et al. 2002; Rout et al. 2006; Chegrouche et al. 2009), people felt threatened by the detection of radioactive materials in drinking water. Ion exchange and reverse osmosis are effective water treatment technologies for removing radioactive materials in general. The removal rate of radioactive material appeared to be over 90% with the ion exchange and reverse osmosis processes (Haberer 1998; Arnal et al. 2003a, b). Table 1 shows the removal efficiency of the unit process of a water treatment plant (Goossens et al. 1989; Gafvert et al. 2002; EPA 2005).

The objective of this study is to evaluate the removal characteristics of radioactive materials in a water treatment process using reference standards of radioactive materials which are stable elements. To evaluate the removal efficiency of reference standards, conventional and advanced processes were investigated. The major processes for evaluation in the laboratory scale study were coagulation/flocculation, activated carbon, zeolites and ion exchange.

**METHODS**

**Tested water**

Surface water (Daechung reservoir, Korea), which is used as a source water for water treatment plants, was used for evaluation purposes to determine removal characterizations of reference standards of radioactive materials. The average annual rates for turbidity and pH of surface water are 2.5 NTU and 7.3, respectively. The turbidity range of the tested period was 0.7–1.1 NTU. As the turbidity of the surface water was relatively low during the tested period, the turbidity was adjusted with river sediments to examine the turbidity effect of the surface water. The adjusted turbidity range of the tested water was 10–100 NTU. Also, dissolved organic carbon in the tested water is 1.3 mg/L.

**Standards/chemicals**

Reference standards used for this study were Cesium-133 (Cs-133, 1,000 μg/mL in 2–5% nitric acid) and Iodide-127 (I-127, 1,000 μg/mL in water). The initial concentrations of Cs-133 and I-127 were 20 μg/L and 100–200 μg/L, respectively. Coagulants used in this test were liquid aluminum sulfate (LAS, 8%), polyaluminum chloride (PAC, 11%), and polyaluminum hydroxychlorosulfate (PAHCS, 12.5%). A jar-test (Daehan Sci., Model DH.CV006010) was used for estimating coagulation and flocculation efficiency. After finding the optimum concentration of coagulants for removal efficiency, powdered activated carbon was evaluated for the removal test. In this test, 20 mg/L of PAC was used as the fixed optimum concentration of coagulants for powdered activated carbon. Cation (DOWEX™ HCR-S/S), anion (DOWEX™ SAR), and mixed bed ion exchange resin (DOWEX™ MONOSPHERE™ MR-450 UPW) were used for the ion-exchange method of removal efficiency. Also, natural and synthetic (WACO, A-4) zeolites were used for this removal test.

<table>
<thead>
<tr>
<th>Overview of water treatment unit processes for removing radioactive materials</th>
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<tbody>
<tr>
<td><strong>Coagulation/Filtration</strong></td>
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<tr>
<td>Gross-alpha</td>
</tr>
<tr>
<td>Gross-beta</td>
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<tr>
<td>Cesium</td>
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<td>Ruthenium</td>
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<td>Iodide</td>
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<td>Uranium</td>
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<td>Plutonium</td>
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O: increased effect of water treatment.
–: No data.
Condition of analysis

The laboratory scale experiment was performed from April to July 2011. Instruments used for the evaluation of removal of reference standards included the inductively coupled plasma–mass spectroscopy (ICP/MS, Perkin Elmer). Using ICP/MS, quality control was performed on Cs-133 and I-127. The detection limits (mg/L) of Cs-133 and I-127 were 0.0004 and 0.0002, respectively. The limits of qualification (mg/L) of Cs-133 and I-127 were 0.0012 and 0.0006, respectively. The degrees of precision (%) of Cs-133 and I-127 were 2.41 and 1.27. Both are less than 5% of the degree of precision. Also, the degrees of accuracy (%) of Cs-133 and I-127 were 99.67 and 94.25, respectively. Therefore, the condition of the instrument and analysis was relatively stable. Also, the linearity of calibration curve ($R^2$) showed 0.9995–1.0. To determine which unit process of water treatment is the most effective to remove reference standards of radioactive materials, conventional and advanced processes were evaluated at laboratory scale. For each process, several conditions, including turbidity (adding kaolin and red soil) and concentration of coagulants, were tested to determine the removal efficiency.

RESULTS AND DISCUSSION

Coagulation/flocculation

The laboratory scale removal evaluation test of reference standards of radioactive materials for each unit process in water treatment was assessed. For evaluating the coagulation and flocculation process, PAC, LAS, and PAHCS were used as coagulants. Also, the turbidity of the sample was prepared as 10, 50, 80, and 100 NTU to investigate the effect of turbidity on the coagulation and flocculation process. The jar-test was used for this experiment.

Figure 1 shows the removal rate of Cs-133 in various concentrations of LAS, PAC, and PAHCS with the variation of turbidity of the water sample. There was no clear trend of removal rate in LAS. Most of the removal rates of Cs-133 were less than 5% in PAC. The higher concentration of 20 mg/L in PAC showed a similar removal rate between concentrations of coagulants. The high-basicity coagulant, PAHCS, showed higher removal rates of Cs-133 compared to the other two coagulants, PAC and LAS. The highest removal rate of Cs-133 occurred in 50 NTU of turbidity in 20 mg/L of PAHCS. The turbidity of 50 NTU showed a higher removal rate in PAC and PAHCS than the other turbidity ranges, which means that a degree of turbidity influences the removal rate of radioactive references.
Figure 2 shows the removal rate of I-127 in various concentrations of LAS, PAC, and PAHCS with the variation of turbidity of water samples. The trends of the removal rate of I-127 in various coagulants with the variation of turbidity were similar to those of Cs-133. The highest removal rate of I-127 in PAHCS occurred in 50 NTU of turbidity in 20 mg/L of PAHCS. However, the removal rate is relatively low in various coagulants with the various turbidity ranges for both Cs-133 and I-127. Therefore, it could be concluded that there is no clear removal efficiency of Cs-133 and I-127 in regard to the coagulation and flocculation process. A previous study on the removal rate of radioactive materials showed relatively higher removal rates compared to this study (Goossens et al. 1989). The condition of the experiment and analysis might be different between these tests.

**Adsorption**

Powdered activated carbon and zeolites were used for the adsorption process of advanced water treatment in order to investigate the removal rates of Cs-133 and I-127. The concentration of coagulant used for the adsorption test was 20 mg/L of PAC as an optimum condition. Cs-133 and I-127 were not removed in 20 mg/L of powdered activated carbon. Also, at 40 mg/L of powdered activated carbon the removal rate was less than 5% (Figure 3). This result suggests that the powdered activated carbon process was not effective in removing radioactive materials. A granular activated carbon process might be useful for removing radioactive materials depending on the operating conditions (non-published K-water data 2011).

In previous studies, the removal kinetics of zeolites and the efficiency of natural zeolites were investigated (Krestou et al. 2003; Han et al. 2007). In this study, natural zeolite and synthetic zeolite were tested for the removal efficiency of reference standards, Cs-133 and I-127. The tested concentrations of both zeolites were 10–300 mg/L (Figure 4). In both zeolites Cs-133 was removed at higher rates than I-127. Synthetic zeolites showed a higher removal rate of Cs-133 than natural zeolites. Also, a higher concentration of zeolites showed higher removal rates of Cs-133. Cs-133 was removed effectively in natural and synthetic zeolites but I-127 was not removed under the same conditions (less than 10%).

**Ion exchange**

Several studies show that the ion exchange process is a useful method for removing radioactive materials (Morton & Straoub 1955; Haberer 1998). In this study, laboratory scale ion exchange was evaluated for the removal of standard references, Cs-133 and I-127. The removal rate of
cation and anion exchange resins showed 99–100 and 80% efficiency for Cs-133 and I-127, respectively (Figure 5). Also, in mixed-bed ion exchange resin, the removal rates were 99% and 92% of Cs-133 and I-127, respectively. The removal rates of the standard references of radioactive material in the ion exchange process were higher than those in the coagulation/flocculation and adsorption processes.

CONCLUSIONS

A laboratory scale removal evaluation test using reference standards of radioactive material for the process of coagulation/flocculation as a conventional water treatment process, and adsorption and ion exchange as advanced water treatment processes was assessed. For coagulation and flocculation, LAS, PAC, and PAHCS were investigated as coagulants for the removal of reference standards of radioactive materials, Cs-133 and I-127. PAHCS showed the highest removal rate in 50 NTU of turbidity. The removal efficiency of coagulants was as follows: PAHCS > PAC > LAS. However, the overall removal rate of coagulation/flocculation for reference standards of radioactive material was low. The powdered activated carbon and zeolites were evaluated as an adsorption process of water treatment. The powdered activated carbon showed no removal at 20 mg/L concentration, and very little at 40 mg/L. Therefore, the powdered activated carbon process appeared to be ineffective in removing the reference radioactive materials. A higher concentration of zeolites showed higher removal rates for Cs-133. However, for I-127, zeolites showed a removal rate of less than 10%. The ion exchange process as an advanced process of water treatment showed higher removal rates for Cs-133 and I-127 compared to the other processes.
treatment was evaluated and shown to be an effective process for removal of both Cs-133 and I-127. In this laboratory scale experiment, the removal of reference standards of radioactive material, and the behavioral characteristics of radioactive materials in a water treatment process could be assessed. Based on these laboratory scale results on the removal of reference standards, the radioactive materials could be characterized and evaluated in water treatment plants in order to supply safe drinking water free from radioactive materials.

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