

## Research Article

# Characterization of Waste Generated from Nuclide Management Process in Waste Burden Minimization Technology for Spent Nuclear Fuel

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To reduce the environmental burden caused by the disposal of spent nuclear fuel, waste burden minimization technology is currently being developed at the Korea Atomic Energy Research Institute. The technology includes a nuclide management process that can maximize disposal efficiency by selectively separating and collecting major nuclides in spent nuclear fuel. To manufacture a waste form of high durability, the characteristics of the waste generated during the process should be evaluated. In this study, the physical, radiological, and thermal characteristics of the waste and waste forms for major nuclides (Cs, Sr, I, transuranic/rare earth, and Tc/Se) generated in the nuclide management process were analyzed. In the case of Cs nuclides, characterization was conducted according to the capture rate of the adsorbent in the high-temperature heat treatment process; meanwhile, in the case of Sr nuclides, characterization was performed by considering the ratio of similar nuclides in the chlorination process. For I nuclide, analysis was performed based on the available waste form, and for TRU/RE and Tc/Se nuclides, analysis was performed by considering chlorination and mid-temperature heat treatment. The radioactivity and heat generation rate of each waste and waste form were evaluated over a period of 1,000 years. The results of this study could be used to derive the centerline temperature for the thermal stability evaluation of waste forms and for the feasibility evaluation of each disposal system considered in the waste burden minimization technology.

## 1. Introduction

To reduce the environmental burden caused by the disposal of spent nuclear fuel, the Korea Atomic Energy Research Institute is currently developing waste burden minimization technology to reduce the disposal area by selectively recovering the main fission products from spent nuclear fuel based on their characteristics, such as long-lived, high-heat-generating, and high-mobility nuclides. The technology comprises a nuclide management process and a waste form

fabrication process. The nuclide management process selectively separates and captures the major nuclides in spent nuclear fuel, thereby maximizing disposal efficiency. Major nuclides, such as Cesium (Cs), strontium (Sr), iodine (I), transuranic/rare earth (TRU/RE), and technetium/selenium (Tc/Se), which are separated based on their characteristics, are fabricated as a waste form while considering their disposal in the waste form fabrication process, where the separated nuclides are to be manufactured as a ceramic waste form of high chemical durability. The final target of the waste

burden minimization technology is to derive the best option for reducing the disposal area by considering various disposal systems to ensure efficiency, safety, and economic feasibility. This could be achieved by combining and optimizing the analysis and test data obtained from the nuclide management and waste form fabrication processes.

The nuclide management process includes a series of processes involving the separation and capture of major nuclides that affect the reduction in the disposal area. A candidate process plan has been established, as shown in Figure 1. The spent nuclear fuel is converted to the  $U_3O_8$  phase through medium-temperature heat treatment between  $500^\circ\text{C}$ – $600^\circ\text{C}$ , where Tc-99 and Se-79 nuclides are volatilized and captured in a Ca-based absorbent [1–3], which is generated as waste to be manufactured in waste form. In the subsequent high-temperature heat treatment process operated between  $1,400^\circ\text{C}$  and  $1,500^\circ\text{C}$  [4], Cs-135/137 and I-129, which are semivolatile and volatile nuclides, respectively, are volatilized and captured by each adsorbent [5–7], which is generated as waste to be manufactured in a waste form. The Sr-90 nuclide, a highly heat-generating nuclide, is separated during the chlorination of  $U_3O_8$  powder after it has melted into a chloride phase and then recovered via the reactive-distillation process [8]. Thereafter, residual salt is removed via a salt removal process, and TRU/RE waste is generated via an additional TRU/RE chlorination process [9–11]. Residual  $U_3O_8$  is manufactured as a  $UO_2$  block after an additional salt removal process. Therefore, the major nuclides of Cs, Sr, I, TRU/RE, and Tc/Se are generated as waste subject to the waste form fabrication process. Prior to manufacturing these waste nuclides into a waste form, it is necessary to predict the characteristics of wastes and waste forms by deriving the fundamental physical properties and radiological/thermal characteristics of each nuclide. This would allow the prediction of the centerline temperature of a waste form according to the size and evaluate the effect on the disposal system.

In this context, the fundamental physical, radiological, and thermal characteristics of wastes and waste forms containing the major nuclides of Cs, Sr, I, TRU/RE, and Tc/Se generated from the nuclide management process were evaluated. The physical form of wastes and waste forms was discussed considering the nuclide management and waste form fabrication processes. The radiological and thermal properties of wastes and waste forms were estimated over 1,000 years. The heat generation and specific activity for each waste form were analyzed in terms of disposal.

## 2. Calculation Method

To evaluate the radioactive properties of waste nuclides generated in the nuclide management process and radiological properties of each waste form (when the waste nuclides are manufactured in the form of ceramic waste), the radioactivity and heat generation characteristics of each waste/waste form were calculated using the Origen program [12]. The nuclide inventory was calculated using PWR Spent Fuel with a Plus-7 library, 4.5 wt% uranium enrichment, 55,000 MWd/MTU burn-up, and 37.5 MW/MTU of average

output. The radioactive decay was calculated using the inventory of each major nuclide after three cycles of operation and 10 years of cooling. Accordingly, the activity (Bq/g) and heat generation ( $\text{W}/\text{m}^3$ ) for the major nuclides were obtained to evaluate the radiological properties of the waste/waste forms over 1,000 years. The waste form with a heat generation rate exceeding  $2\text{ kW}/\text{m}^3$  and an alpha-activity exceeding 4,000 Bq/g was classified as high-level waste based on the current classification criteria of the Republic of Korea, where the alpha activity indicates the activity caused by an  $\alpha$ -emitting nuclide with a half-life exceeding 20 years. To confirm the waste type, the alpha activity (alpha-Bq/g) was calculated using all actinides and some rare earth (RE) elements (Nd-144, Sm-146, 147, 148, 149, Gd-152) emitting alpha nuclides with a half-life exceeding 20 years.

The radiological characteristics of the waste/waste forms were evaluated using density. High density results in a conservative radiological evaluation; therefore, the density of each waste and waste form was set based on the theoretical density of the corresponding phase. The waste loadings of the waste forms were evaluated based on the nuclide inventory and density. The waste loadings were determined based on the weight of the nuclide in its waste form. Subsequently, the properties of the wastes and waste forms were predicted while considering the characteristics of the nuclide management and the waste form fabrication processes, respectively.

## 3. Results and Discussion

**3.1. Characterization of Cs Waste/Waste Form.** Cs nuclides are volatilized during the high-temperature heat treatment process of the nuclide management process and then captured using aluminosilicate-based adsorbents, which are primarily composed of  $Al_6Si_2O_{13}$  (mullite) +  $4SiO_2$  [5, 6]. Cs nuclides are captured in the form of Cs-pollucite ( $CsAlSi_2O_6$ ) and generated as waste [13], which is subsequently fabricated as a ceramic waste in the form of Cs-pollucite by pelletizing and sintering the waste. Therefore, in the case of Cs nuclides, the waste and waste forms have the same phase, and they basically show the same radiological properties. However, considering the capturing process, the amount of Cs nuclides in the adsorbent is expected to differ based on the capture rate of the adsorbent nuclides. Consequently, the radiological properties of the waste/waste form were evaluated according to the capture rate of Cs nuclides (conversion rate of Cs-pollucite) based on the weight of pollucite in the adsorbent.

To calculate the density of each phase, the theoretical densities of pollucite ( $2.9\text{ g}/\text{cm}^3$ ), mullite ( $3.0\text{ g}/\text{cm}^3$ ), and  $SiO_2$  ( $2.648\text{ g}/\text{cm}^3$ ) were used, and the density of the waste/waste form was evaluated by considering the weight ratio based on the capture rate. Table 1 shows the densities and resulting waste loadings of the Cs waste/waste form based on the expected capture rate. In the case of a 100% capture rate, a high waste loading of 42.6 wt% is expected.

The radiological properties of the Cs waste/waste form were calculated and are presented in Table 2 and Figure 2. The activity of Cs waste/waste form was proportional to the capture rate and exhibited a high initial activity level of  $10^{11}$ -

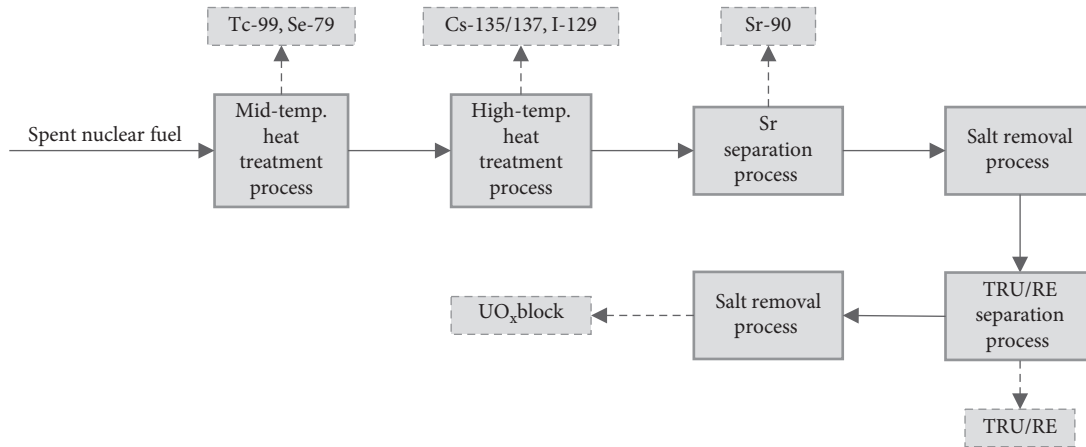


FIGURE 1: Schematic of nuclide management process and major nuclides generated from each process.

TABLE 1: Densities and waste loadings of Cs waste/waste form according to capture rate of Cs nuclide.

Capture rate	25%	50%	75%	100%
Density (g/cm <sup>3</sup> )	2.872	2.881	2.889	2.900
Waste loading	10.6%	21.3%	29.8%	42.6%

TABLE 2: Radiological characteristics of Cs waste/waste form according to capture rate.

Capture rate	Year							
	0.3	1	3	10	30	100	300	1000
	Activity (Bq/g)							
25%	2.676E+11	2.616E+11	2.466E+11	2.071E+11	1.302E+11	2.583E+10	2.549E+08	7.542E+05
50%	5.352E+11	5.232E+11	4.932E+11	4.142E+11	2.604E+11	5.166E+10	5.098E+08	1.508E+06
75%	7.492E+11	7.325E+11	6.905E+11	5.799E+11	3.646E+11	7.233E+10	7.137E+08	2.112E+06
100%	1.070E+12	1.046E+12	9.864E+11	8.284E+11	5.209E+11	1.033E+11	1.020E+09	3.017E+06
	Heat generation (W/m <sup>3</sup> )							
25%	5.688E+04	5.458E+04	4.963E+04	4.009E+04	2.508E+04	4.975E+03	4.896E+01	1.955E-02
50%	1.141E+05	1.095E+05	9.958E+04	8.044E+04	5.031E+04	9.983E+03	9.823E+01	3.922E-02
75%	1.602E+05	1.537E+05	1.398E+05	1.129E+05	7.062E+04	1.401E+04	1.379E+02	5.505E-02
100%	2.297E+05	2.205E+05	2.005E+05	1.619E+05	1.013E+05	2.010E+04	1.977E+02	7.895E-02

<sup>12</sup>Bq/g. No alpha activity occurred due to the absence of actinides or RE nuclides. Owing to the high-heat-generating properties of Cs nuclides (initial specific heat generation rate of  $1.860 \times 10^{-1}$  W/g), the Cs waste/waste form indicated a high initial heat generation rate. When the capture rate was 100%, the heat generation rate became  $2.3 \times 10^5$  W/m<sup>3</sup>, which exceeded the heat generation rate of a high-level waste; however, it could not be classified as that of high-level waste due to the absence of alpha-emitting nuclides. When the capture rate was 25%, the heat generation rate decreased slightly to approximately  $5.7 \times 10^4$  W/m<sup>3</sup>, although the Cs waste/waste form exhibited high exothermic properties. Consequently, particular care needs to be taken with regard to handling or storage of Cs waste/waste form. The obtained thermal characteristics will be used as preliminary data for determining the waste form size and thermal stability analysis through the calculation of the centerline temperature.

**3.2. Characterization of Sr Waste/Waste Form.** Sr-90 nuclide, a highly heat-generating nuclide, is separated during the chlorination process in the nuclide management process. Spent nuclear fuel powder in the form of U<sub>3</sub>O<sub>8</sub>, which has undergone mid- and high-temperature heat treatment, is immersed in molten salt, where the Sr nuclides are separated by dissolution in the molten salt. At this time, the Ba nuclides can be dissolved owing to their chemical similarity with the Sr nuclides. Subsequently, the dissolved Sr and Ba nuclides are separated into a precipitated phase via carbonate precipitation, and the residual salt is removed via distillation process. A mixture of Sr/Ba nuclides is generated as waste and is expected to be recovered in an oxide form. The ratio of Sr and Ba nuclides resulting from the separation process can be determined based on the difference in solubility by varying the process conditions. Therefore, in this characterization, the radiological characteristics of the waste were evaluated by varying the ratio of Sr/Ba while

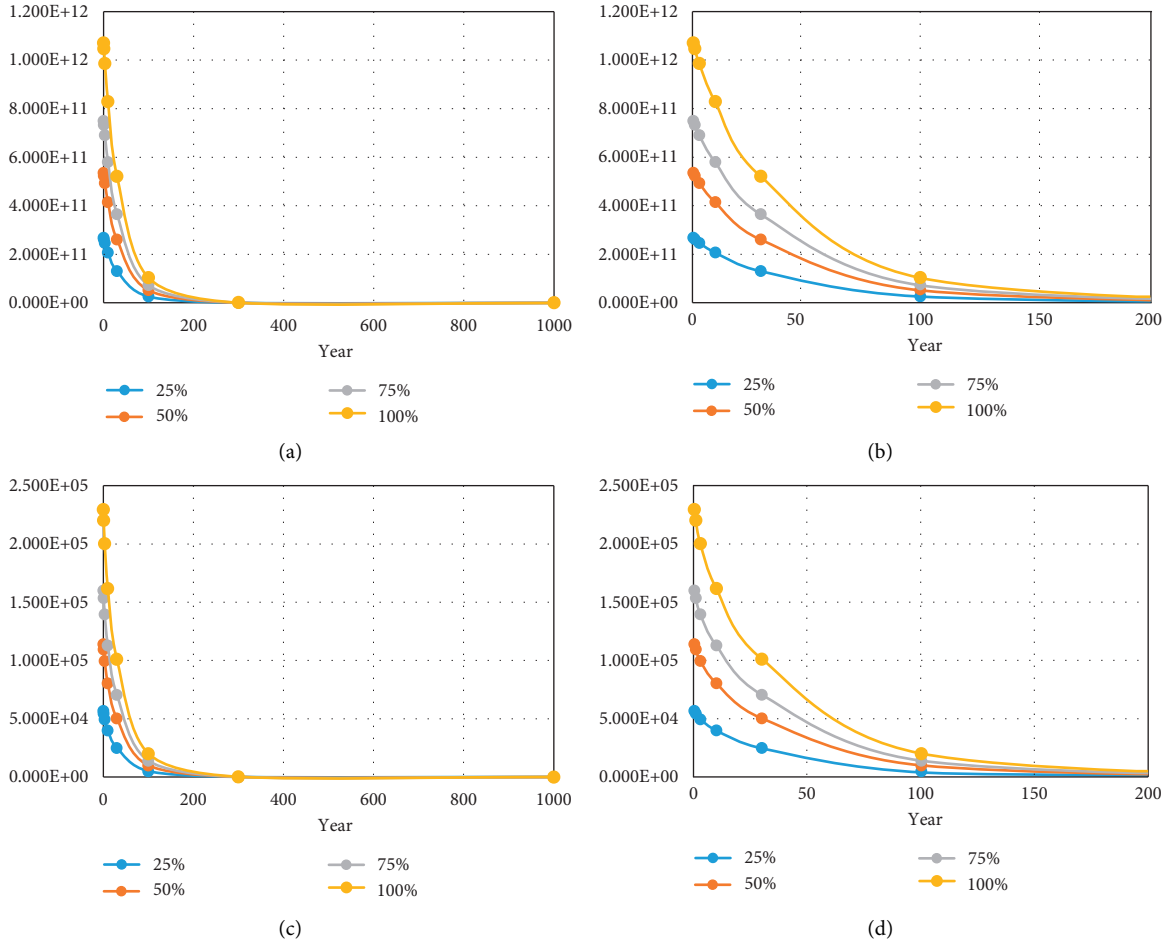


FIGURE 2: Radiological characteristics of Cs waste/waste form according to capture rate ((b) and (d) correspond to magnified graphs of (a) and (c), respectively).

considering all possibilities for the process variables. The composition of the waste was set to  $\text{SrO}$ ,  $\text{Sr}_{0.8}\text{Ba}_{0.2}\text{O}$ ,  $\text{Sr}_{0.6}\text{Ba}_{0.4}\text{O}$ ,  $\text{Sr}_{0.5}\text{Ba}_{0.5}\text{O}$ ,  $\text{Sr}_{0.4}\text{Ba}_{0.6}\text{O}$ ,  $\text{Sr}_{0.2}\text{Ba}_{0.8}\text{O}$ , and  $\text{BaO}$ , assuming that it was separated into pure oxides. The density was calculated as shown in Table 3 using the densities of  $\text{SrO}$  ( $5.009 \text{ g/cm}^3$ ) and  $\text{BaO}$  ( $5.720 \text{ g/cm}^3$ ), based on the composition.

Table 4 and Figure 3 show the radiological properties calculated based on the Sr/Ba ratios in the Sr nuclide waste. Owing to the difference in the initial specific heat generation rate of each nuclide (Sr:  $5.142 \times 10^{-1} \text{ W/g}$ , Ba:  $5.860 \times 10^{-8} \text{ W/g}$ ), the radiological properties of the Sr nuclide waste were dominated by the Sr nuclide. In the case of pure  $\text{BaO}$ , the initial heat generation was approximately  $10^{-1} \text{ W/m}^3$ ; however, when Sr nuclides of  $\text{Sr}_{0.2}\text{Ba}_{0.8}\text{O}$  or higher Sr nuclides were included, it increased rapidly to approximately  $10^{5-6} \text{ W/m}^3$ . Based on the characteristic data, the design of the nuclide management and waste fabrication processes should consider the high activity and exothermic properties of the Sr nuclides.

Sr nuclide waste is planned to be fabricated in a waste form using a mineral system showing natural analogy. Therefore, it is desirable to be manufactured as a  $\text{SrTiO}_3$

phase, which is one of the phases of the Synroc nuclear waste form [14]. The equal molar equivalents of the  $\text{TiO}_2$  matrix and SrO waste, generated in the nuclide management process, were mixed to produce  $\text{SrTiO}_3$  via the calcination/sintering process. Considering the characteristics of the nuclide management process, the representative waste forms to be manufactured were in the phase of  $\text{SrTiO}_3$ ,  $\text{Sr}_{0.8}\text{Ba}_{0.2}\text{TiO}_3$ ,  $\text{Sr}_{0.6}\text{Ba}_{0.4}\text{TiO}_3$ ,  $\text{Sr}_{0.5}\text{Ba}_{0.5}\text{TiO}_3$ ,  $\text{Sr}_{0.4}\text{Ba}_{0.6}\text{TiO}_3$ ,  $\text{Sr}_{0.2}\text{Ba}_{0.8}\text{TiO}_3$ , and  $\text{BaTiO}_3$  according to the Sr/Ba ratios.

To estimate the density of the Sr waste form for each composition, the density was linearly fitted, for each Sr/Ba ratio, using values reported in the literature [15]. The resulting waste loadings are presented in Table 3. As the amount of the relatively heavy Ba nuclides in  $\text{SrTiO}_3$  increases, the density increases (approximately from 5 to  $6 \text{ g/cm}^3$ ), which is expected to increase the waste loading from 47.8% to 58.9%.

The radiological characteristics of the Sr waste form, based on the Sr/Ba ratios, were analyzed using the density and waste loading values and are shown in Table 5 and Figure 4. Similar to the Sr nuclide waste, the radiological properties of the Sr waste form were governed by the content of Sr nuclides, which exhibited high specific radioactivity and a high heat generation rate. Hence, high specific

TABLE 3: Densities and waste loadings of Sr waste/waste form according to Sr/Ba ratio.

Waste	Density (g/cm <sup>3</sup> )	Waste form	Density (g/cm <sup>3</sup> )	Waste loading (%)
SrO	5.009	SrTiO <sub>3</sub>	5.08	47.8
Sr <sub>0.8</sub> Ba <sub>0.2</sub> O	5.183	Sr <sub>0.8</sub> Ba <sub>0.2</sub> TiO <sub>3</sub>	5.27	50.4
Sr <sub>0.6</sub> Ba <sub>0.4</sub> O	5.339	Sr <sub>0.6</sub> Ba <sub>0.4</sub> TiO <sub>3</sub>	5.46	52.9
Sr <sub>0.5</sub> Ba <sub>0.5</sub> O	5.410	Sr <sub>0.5</sub> Ba <sub>0.5</sub> TiO <sub>3</sub>	5.56	54.0
Sr <sub>0.4</sub> Ba <sub>0.6</sub> O	5.478	Sr <sub>0.4</sub> Ba <sub>0.6</sub> TiO <sub>3</sub>	5.65	55.1
Sr <sub>0.2</sub> Ba <sub>0.8</sub> O	5.605	Sr <sub>0.2</sub> Ba <sub>0.8</sub> TiO <sub>3</sub>	5.84	57.1
BaO	5.720	BaTiO <sub>3</sub>	6.03	58.9

TABLE 4: Radiological characteristics of Sr waste according to Sr/Ba ratio.

Year	0.3	1	3	10	30	100	300	1000
	Activity (Bq/g)							
SrO	4.801E+12	4.719E+12	4.493E+12	3.781E+12	2.311E+12	4.122E+11	2.993E+09	9.767E+01
Sr <sub>0.8</sub> Ba <sub>0.2</sub> O	3.505E+12	3.445E+12	3.279E+12	2.760E+12	1.687E+12	3.009E+11	2.185E+09	7.129E+01
Sr <sub>0.6</sub> Ba <sub>0.4</sub> O	2.417E+12	2.376E+12	2.262E+12	1.904E+12	1.163E+12	2.075E+11	1.507E+09	4.916E+01
Sr <sub>0.5</sub> Ba <sub>0.5</sub> O	1.936E+12	1.903E+12	1.812E+12	1.525E+12	9.320E+11	1.662E+11	1.207E+09	3.939E+01
Sr <sub>0.4</sub> Ba <sub>0.6</sub> O	1.491E+12	1.466E+12	1.395E+12	1.175E+12	7.178E+11	1.280E+11	9.297E+08	3.033E+01
Sr <sub>0.2</sub> Ba <sub>0.8</sub> O	6.939E+11	6.821E+11	6.493E+11	5.465E+11	3.340E+11	5.958E+10	4.326E+08	1.412E+01
BaO	7.192E+05	6.868E+05	6.020E+05	3.796E+05	1.016E+05	1.009E+03	1.906E-03	1.769E-23
	Heat generation (W/m <sup>3</sup> )							
SrO	2.178E+06	2.141E+06	2.038E+06	1.715E+06	1.048E+06	1.870E+05	1.357E+03	4.430E-05
Sr <sub>0.8</sub> Ba <sub>0.2</sub> O	1.645E+06	1.617E+06	1.539E+06	1.295E+06	7.915E+05	1.412E+05	1.025E+03	3.346E-05
Sr <sub>0.6</sub> Ba <sub>0.4</sub> O	1.168E+06	1.148E+06	1.093E+06	9.201E+05	5.622E+05	1.003E+05	7.283E+02	2.377E-05
Sr <sub>0.5</sub> Ba <sub>0.5</sub> O	9.487E+05	9.324E+05	8.876E+05	7.470E+05	4.564E+05	8.143E+04	5.913E+02	1.930E-05
Sr <sub>0.4</sub> Ba <sub>0.6</sub> O	7.399E+05	7.272E+05	6.922E+05	5.826E+05	3.560E+05	6.351E+04	4.612E+02	1.505E-05
Sr <sub>0.2</sub> Ba <sub>0.8</sub> O	3.522E+05	3.462E+05	3.296E+05	2.774E+05	1.695E+05	3.024E+04	2.195E+02	7.165E-06
BaO	3.002E-01	2.866E-01	2.512E-01	1.584E-01	4.241E-02	4.209E-04	7.956E-10	7.382E-30

radioactivity ( $\sim 10^{11-12}$  Bq/g) and high heat generation rate ( $\sim 10^{5-6}$  W/m<sup>3</sup>) were obtained even when a small quantity of Sr nuclides was included in the waste form. The Sr waste form is expected to exhibit a high heat generation rate; therefore, the centerline temperature of the waste form can be increased based on the waste loading, which may adversely affect the thermal durability. Consequently, the aforementioned results can be applied to estimate the centerline temperature in conjunction with the experimentally obtained thermal conductivity data of the waste form [16]. If the estimated thermal integrity is not to be within the range of the durability limit, the waste loading must be limited. This could be done by adding a dummy material such as natural Sr, in the waste form, to reduce the heat generation rate during the waste form fabrication process.

**3.3. Characterization of I Waste/Waste Form.** I nuclides are volatilized in the high-temperature heat treatment process of the nuclide management process and then collected in the form of AgI using a silica aerogel-based adsorbent. Because the Si-based adsorbent does not exhibit a natural analogy for the mineralization of AgI, it is planned to be separated into AgI via a distillation/recovery process [17] and then manufactured as an I waste form. Therefore, during the nuclide management process, I nuclide waste was generated in the AgI phase with a density of 5.69 g/cm<sup>3</sup>. The radiological

properties of AgI were evaluated and are presented in Table 6 and Figure 5. Because the I-129 nuclide has a half-life that exceeds 15 million years, the I nuclide waste did not change over 1,000 years. This indicated extremely low radioactivity and heat generation; therefore, a high level of caution is not necessary during the waste form fabrication process.

The AgI waste is planned to be fabricated in a ceramic or glass waste form. Considering natural analogy, apatite is a good choice for the ceramic waste form [18]. In particular, iodoapatite (Pb<sub>10</sub>(VO<sub>4</sub>)<sub>6</sub>I<sub>2</sub>) was selected as a mineral system that could accommodate I nuclides in the crystal structure. The density of iodoapatite is 6.8 g/cm<sup>3</sup>, and because it forms a crystal structure with a fixed composition, the waste loading was fixed at 8.4 wt% (based on iodine). Meanwhile, the radiological analysis was also performed on the iodine glass waste form. Recently, this waste form has been actively researched to find ways to solidify the I nuclide. Notably, Ag<sub>2</sub>O-AgI-M<sub>2</sub>O<sub>3</sub>, where M is a metal cation, is considered because the glass system can accommodate I nuclides with high waste loading compared to the iodoapatite ceramic waste form [19–21]. Since the composition could be diversified from the glass system, the waste loading was not fixed. Therefore, in the case of the iodine glass waste form, radiological characteristic analysis was performed according to the waste loading of AgI. The density of the iodine glass waste form was conservatively set as 6.5 g/cm<sup>3</sup> [22], and a characteristic analysis was performed by varying the waste loading to 10, 30, and 50 wt%.

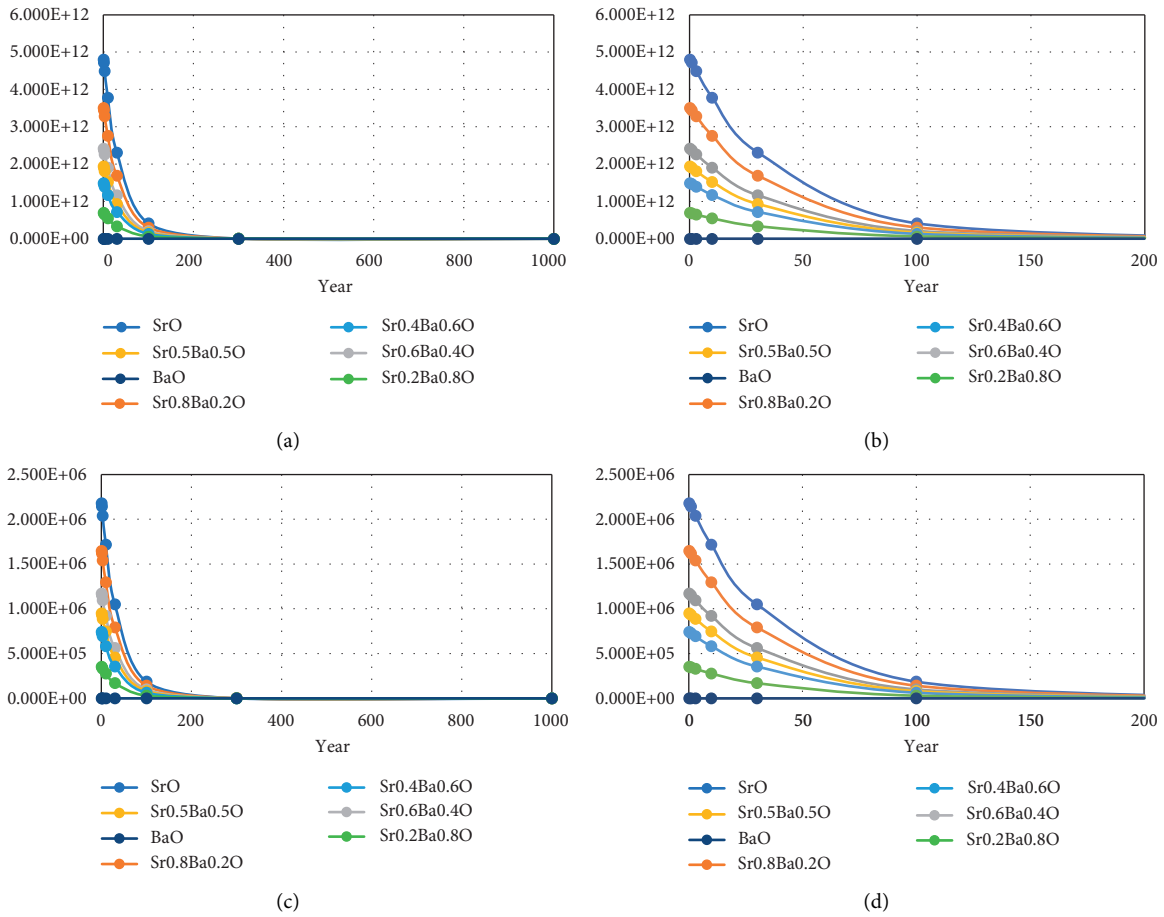


FIGURE 3: Radiological characteristics of Sr waste according to Sr/Ba ratio ((b) and (d) correspond to magnified graphs of (a) and (c), respectively).

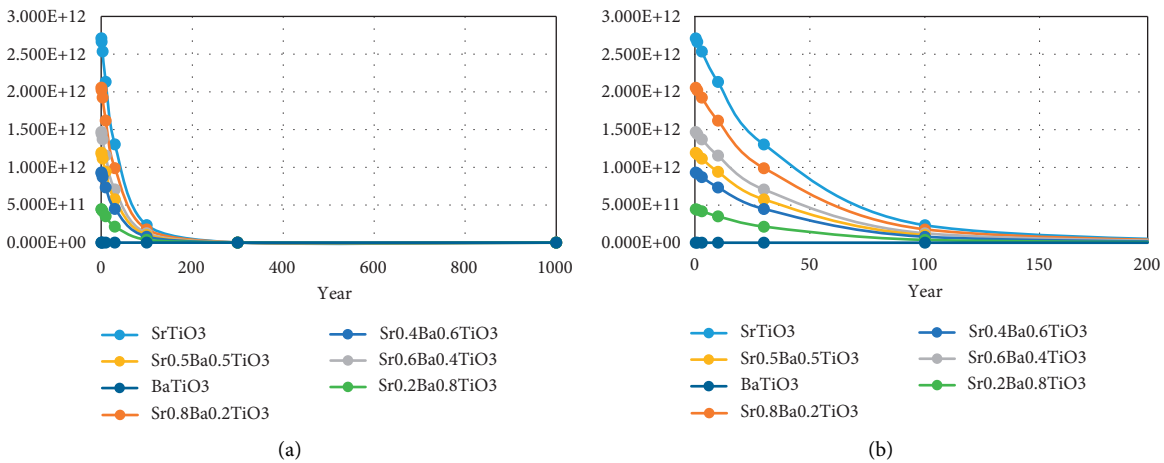


FIGURE 4: Continued.

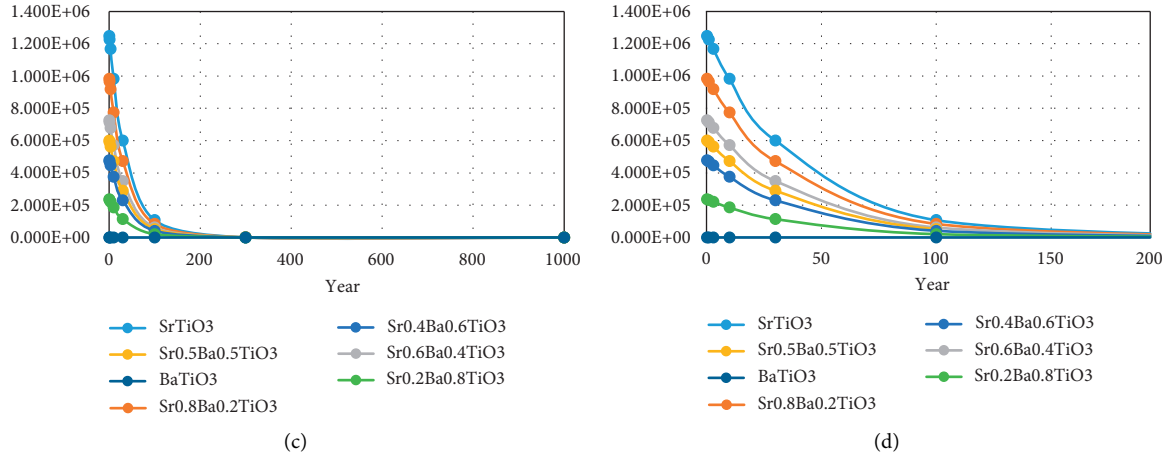


FIGURE 4: Radiological characteristics of Sr waste form according to Sr/Ba ratio ((b) and (d) correspond to magnified graphs of (a) and (c), respectively).

TABLE 5: Radiological characteristics of Sr waste form according to Sr/Ba ratio.

Year	0.3	1	3	10	30	100	300	1000
Activity (Bq/g)								
SrTiO <sub>3</sub>	2.711E+12	2.665E+12	2.537E+12	2.136E+12	1.305E+12	2.328E+11	1.690E+09	5.515E+01
Sr <sub>0.8</sub> Ba <sub>0.2</sub> TiO <sub>3</sub>	2.058E+12	2.022E+12	1.925E+12	1.621E+12	9.904E+11	1.767E+11	1.283E+09	4.186E+01
Sr <sub>0.6</sub> Ba <sub>0.4</sub> TiO <sub>3</sub>	1.468E+12	1.443E+12	1.373E+12	1.156E+12	7.065E+11	1.260E+11	9.151E+08	2.986E+01
Sr <sub>0.5</sub> Ba <sub>0.5</sub> TiO <sub>3</sub>	1.194E+12	1.174E+12	1.117E+12	9.404E+11	5.747E+11	1.025E+11	7.444E+08	2.429E+01
Sr <sub>0.4</sub> Ba <sub>0.6</sub> TiO <sub>3</sub>	9.329E+11	9.170E+11	8.729E+11	7.348E+11	4.490E+11	8.010E+10	5.816E+08	1.898E+01
Sr <sub>0.2</sub> Ba <sub>0.8</sub> TiO <sub>3</sub>	4.457E+11	4.381E+11	4.170E+11	3.510E+11	2.145E+11	3.827E+10	2.779E+08	9.066E+00
BaTiO <sub>3</sub>	4.729E+05	4.516E+05	3.958E+05	2.496E+05	6.678E+04	6.631E+02	1.253E-03	1.163E-23
Heat generation (W/m <sup>3</sup> )								
SrTiO <sub>3</sub>	1.247E+06	1.226E+06	1.167E+06	9.822E+05	6.002E+05	1.071E+05	7.775E+02	2.537E-05
Sr <sub>0.8</sub> Ba <sub>0.2</sub> TiO <sub>3</sub>	9.820E+05	9.652E+05	9.188E+05	7.733E+05	4.725E+05	8.430E+04	6.121E+02	1.998E-05
Sr <sub>0.6</sub> Ba <sub>0.4</sub> TiO <sub>3</sub>	7.258E+05	7.133E+05	6.790E+05	5.715E+05	3.492E+05	6.230E+04	4.524E+02	1.476E-05
Sr <sub>0.5</sub> Ba <sub>0.5</sub> TiO <sub>3</sub>	6.006E+05	5.904E+05	5.620E+05	4.730E+05	2.890E+05	5.156E+04	3.744E+02	1.222E-05
Sr <sub>0.4</sub> Ba <sub>0.6</sub> TiO <sub>3</sub>	4.773E+05	4.692E+05	4.466E+05	3.759E+05	2.297E+05	4.098E+04	2.975E+02	9.710E-06
Sr <sub>0.2</sub> Ba <sub>0.8</sub> TiO <sub>3</sub>	2.357E+05	2.317E+05	2.205E+05	1.856E+05	1.134E+05	2.023E+04	1.469E+02	4.795E-06
BaTiO <sub>3</sub>	2.081E-01	1.987E-01	1.741E-01	1.098E-01	2.940E-02	2.918E-04	5.515E-10	5.117E-30

TABLE 6: Radiological characteristics of I waste.

Year	0.3	1	3	10	30	100	300	1000
Bq/g	2.723E+06	2.723E+06	2.723E+06	2.723E+06	2.723E+06	2.723E+06	2.723E+06	2.723E+06
W/m <sup>3</sup>	1.959E-01	1.959E-01	1.959E-01	1.959E-01	1.959E-01	1.959E-01	1.959E-01	1.959E-01

Table 7 and Figure 6(a) show the radiological characteristics of the iodapatite waste form. Table 8 and Figure 6(b) show the radiological characteristics of the iodine glass waste form based on the waste loading. Both waste forms showed low radioactivity and heat generation rates due to I-129, a long-lived nuclide. Moreover, the iodine glass waste form containing AgI-10wt%, which was similar to the waste loading of iodapatite, showed radiological properties similar to those of the iodapatite waste form. Consequently, increasing the waste loading due to low radioactivity and heat generation could be advantageous in terms of economy and waste volume reduction. In this respect, the iodine glass

waste form is more advantageous than the iodapatite waste form.

3.4. Characterization of TRU/RE Waste/Waste Form. In the nuclide management process, after the Sr nuclides chlorination process, TRU/RE chlorination is performed to separate TRU and RE nuclides from the U<sub>3</sub>O<sub>8</sub> powder. In the TRU/RE chlorination process, separation is performed through chlorination using oxidizing agents such as UCl<sub>3</sub>, ZrCl<sub>4</sub>, and NH<sub>4</sub>Cl, where TRU and RE nuclides are separated simultaneously owing to their chemical similarity.

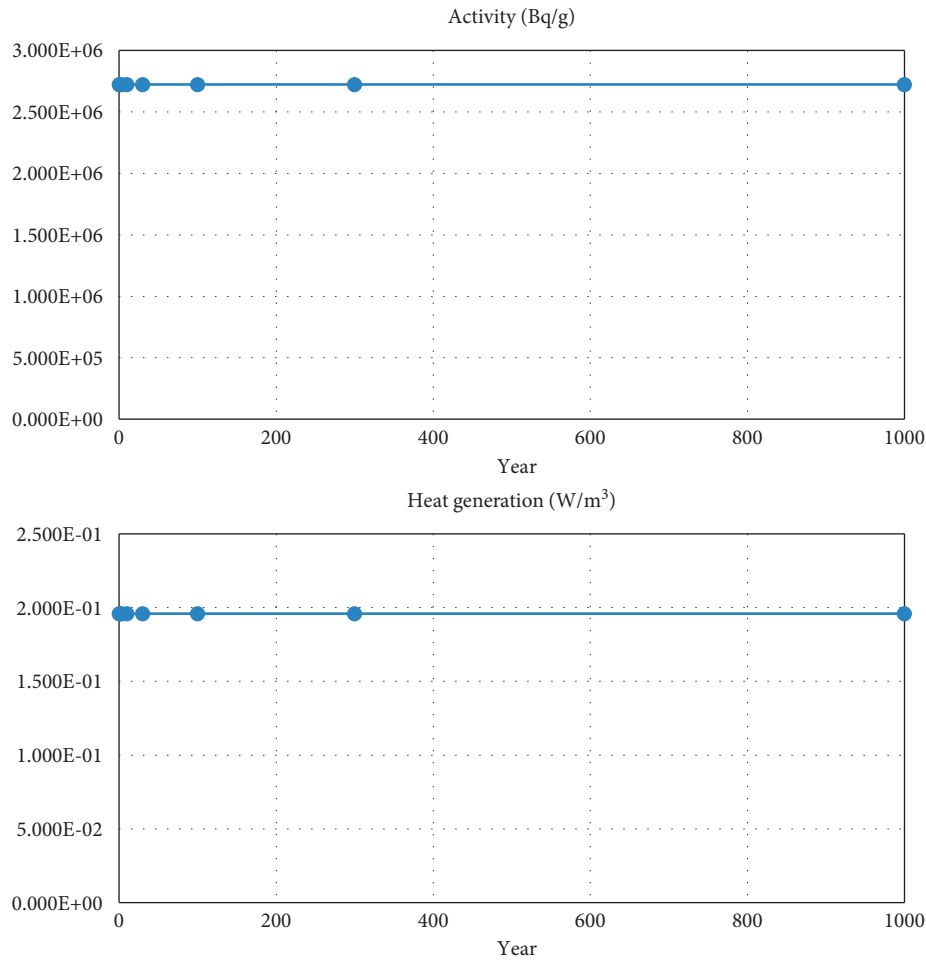


FIGURE 5: Radiological characteristics of I waste.

TABLE 7: Radiological characteristics of iodoapatite waste form.

Year	0.3	1	3	10	30	100	300	1000
Bq/g	$4.240E+05$	$4.240E+05$	$4.240E+05$	$4.240E+05$	$4.240E+05$	$4.240E+05$	$4.240E+05$	$4.240E+05$
W/m <sup>3</sup>	$3.646E-02$	$3.646E-02$	$3.646E-02$	$3.646E-02$	$3.646E-02$	$3.646E-02$	$3.646E-02$	$3.646E-02$

Chlorinated TRU/RE nuclides are separated from salts via distillation after precipitation using a phosphate precipitation agent [9–11]. Therefore, the separated TRU/RE nuclide waste comprises (TRU, RE)PO<sub>4</sub> phases, representing a monazite mineral system with excellent chemical and radiation stability. Additionally, the TRU/RE nuclide waste is sintered in a cylindrical form, without calcination, to yield a TRU/RE waste form [23]. Because the TRU/RE nuclide waste and waste form have the same phase, they exhibit the same radiological properties. Table 9 shows the total amount and ratio of TRU and RE nuclides contained in 10 tons of spent nuclear fuel. (TRU, RE)PO<sub>4</sub> has a density of 5.441 g/cm<sup>3</sup>, and the waste loading considering the amount of TRU/RE nuclide was estimated to be 64.4%.

Table 10 and Figure 7 show the results of the radiological characterization of the TRU/RE nuclide waste/waste form. High initial activity ( $\sim 10^{11}$  Bq/g) and high heat generation

rates ( $75 \text{ kW/m}^3$ ) were indicated, owing to the TRU nuclides. Unlike other waste forms, TRU/RE waste/waste form contains alpha-emitting nuclides (all actinides and some REs, such as Nd-144, Sm-146, 147, 148, 149, and Gd-152), indicating high alpha activity ( $\sim 10^{11}$  Bq/g). The TRU/RE waste/waste form can be classified as high-level waste, as the values of its properties exceed the high-level waste classification criteria (i.e., heat generation rate  $> 2 \text{ kW/m}^3$  and radioactivity concentration  $> 4,000 \alpha\text{-Bq/g}$ ) [24]. In addition, due to the long half-life of TRU nuclides, it is expected to exceed the criteria within 1,000 years. In the case of TRU/RE waste/waste form, it is necessary to establish the nuclide management and the waste form fabrication processes that comply with the high-level waste handling standards. Moreover, the stability should be secured by conducting thermal characteristic evaluations of the waste form in consideration of exothermic characteristics.



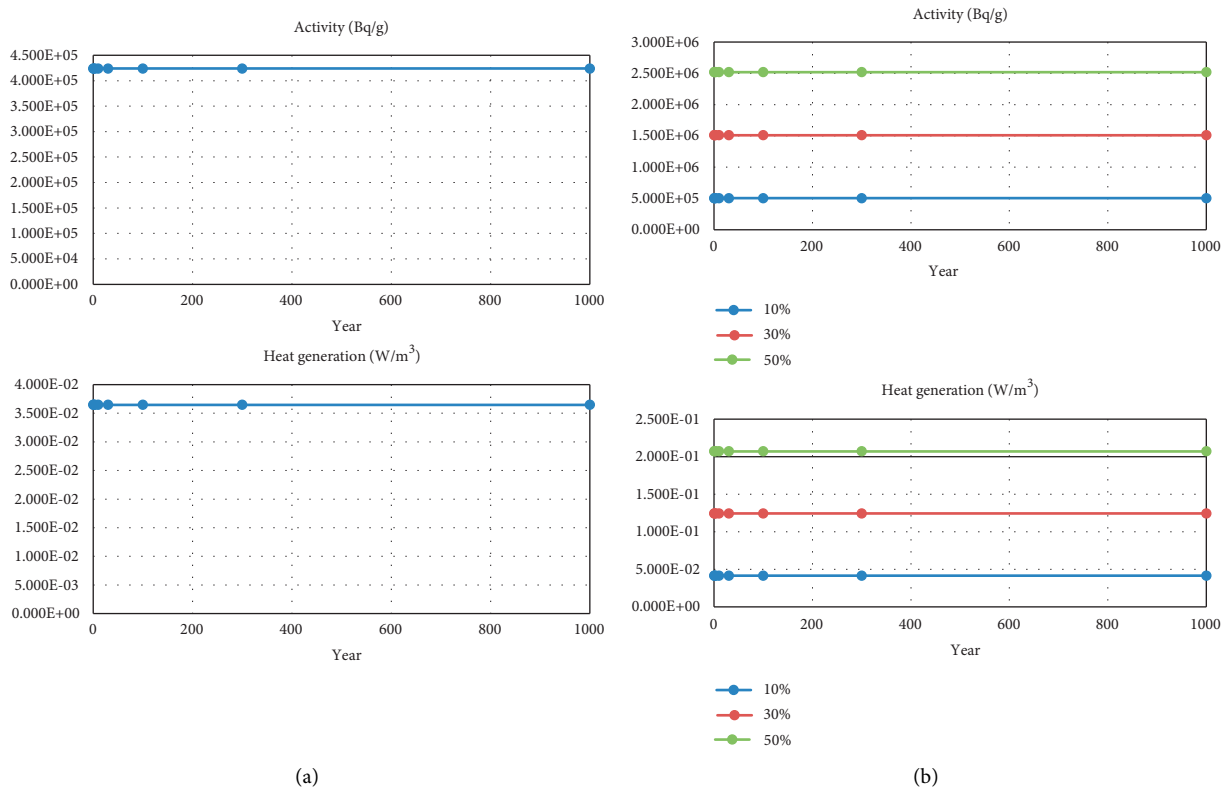


FIGURE 6: Radiological characteristics of I waste form. (a) Iodoapatite waste form and (b) Iodine glass waste form according to waste loading.

TABLE 8: Radiological characteristics of iodine glass waste form according to waste loading.

Year	0.3	1	3	10	30	100	300	1000
	Activity (Bq/g)							
AgI-10%	5.038E+05	5.038E+05	5.038E+05	5.038E+05	5.038E+05	5.038E+05	5.038E+05	5.038E+05
AgI-30%	1.511E+06	1.511E+06	1.511E+06	1.511E+06	1.511E+06	1.511E+06	1.511E+06	1.511E+06
AgI-50%	2.519E+06	2.519E+06	2.519E+06	2.519E+06	2.519E+06	2.519E+06	2.519E+06	2.519E+06
	Heat generation (W/m³)							
AgI-10%	4.141E-02	4.141E-02	4.141E-02	4.141E-02	4.141E-02	4.141E-02	4.141E-02	4.141E-02
AgI-30%	1.242E-01	1.242E-01	1.242E-01	1.242E-01	1.242E-01	1.242E-01	1.242E-01	1.242E-01
AgI-50%	2.070E-01	2.070E-01	2.070E-01	2.070E-01	2.070E-01	2.070E-01	2.070E-01	2.070E-01

3.5. *Characterization of Tc/Se Waste/Waste Form.* In the mid-temperature heat treatment of the nuclide management process, Tc/Se nuclides volatilize and are collected using a CaO-based adsorbent (Figure 1). Subsequently, the Tc/Se nuclides react chemically with the CaO adsorbent to be converted into  $\text{Ca}_5\text{Tc}_2\text{O}_{12}$  or  $\text{Ca}(\text{TcO}_4)_2$  phase and the  $\text{CaSeO}_3$  phase, respectively, to generate waste for waste form fabrication [1–3]. A mineral system with a natural analogy to Tc and Se nuclides does not exist. Therefore, if durability is secured after the physical/chemical/thermal characterization of the waste generated in the nuclide management process, the waste is manufactured and managed as waste form. If durability is not secured, the waste should be manufactured as a waste form that can secure durability using other binders or matrices. Therefore, at the current stage, the radiological characterization of Tc/Se waste was conducted by assuming that the captured phase was a waste/waste form. In the case

of calcium selenite ( $\text{CaSeO}_3$ ), a waste loading of 47.3% was observed, and the density was not reported; therefore, the analysis was performed based on  $3.796 \text{ g/cm}^3$ , i.e., the density of a similar phase, calcium selenite ( $\text{CaSe}_{1.5}\text{O}_4$ ). Table 11 and Figure 8 show the radiological characteristics of the Se nuclide waste/waste form. The Se-79 nuclide is a long-lived nuclide with a half-life of 327,000 years, and its initial radioactivity and heat generation rate were evaluated to be extremely low, i.e.,  $\sim 10^7 \text{ Bq/g}$  and  $10^{-1} \text{ W/m}^3$ , respectively. However, as Se is a highly mobile nuclide [25, 26], a highly durable waste form that can prevent the leakage of nuclides must be developed.

Tc nuclide reacts chemically with the CaO adsorbent generating a waste of  $\text{Ca}_5\text{Tc}_2\text{O}_{12}$  or  $\text{Ca}(\text{TcO}_4)_2$ . However, owing to the absence of the physical properties of the phase, characterization was performed using  $\text{Ca}(\text{ReO}_4)_2$  and  $\text{Ca}_5\text{Re}_2\text{O}_{12}$  phases (as Re is a surrogate of the Tc

TABLE 9: Amount of TRU/RE nuclide contained in 10 tons of spent nuclear fuel.

	Weight (g)	Ratio (%)
Ac	$1.397E-06$	0.00
Th	$1.008E-01$	0.00
Pa	$8.075E-03$	0.00
U	$0.000E+00$	0.00
Np	$8.360E+03$	2.72
Pu	$1.165E+05$	37.86
Am	$1.060E+04$	3.44
Cm	$9.765E+02$	0.32
Bk	$5.841E-08$	0.00
Cf	$2.623E-04$	0.00
Y	$7.296E+03$	2.37
La	$2.012E+04$	6.54
Ce	$3.954E+04$	12.86
Pr	$1.839E+04$	5.98
Nd	$6.707E+04$	21.80
Pm	$1.527E+02$	0.05
Sm	$1.358E+04$	4.42
Eu	$2.273E+03$	0.74
Gd	$2.712E+03$	0.88
Tb	$4.357E+01$	0.01
Dy	$2.249E+01$	0.01
Ho	$1.560E+00$	0.00
Er	$6.005E-01$	0.00
Tm	$4.885E-03$	0.00
Yb	$1.030E-02$	0.00
Total	$3.076E+05$	100.00

TABLE 10: Radiological characteristics of TRU/RE waste/waste form.

Year	0.3	1	3	10	30	100	300	1000
Bq/g	$1.188E+11$	$1.136E+11$	$1.011E+11$	$7.187E+10$	$3.299E+10$	$8.693E+09$	$4.626E+09$	$1.905E+09$
Alpha-Bq/g	$1.031E+11$	$1.000E+11$	$9.183E+10$	$6.861E+10$	$3.226E+10$	$8.500E+09$	$4.584E+09$	$1.905E+09$
W/m <sup>3</sup>	$7.478E+04$	$7.388E+04$	$7.162E+04$	$6.545E+04$	$5.430E+04$	$3.757E+04$	$2.198E+04$	$8.883E+03$

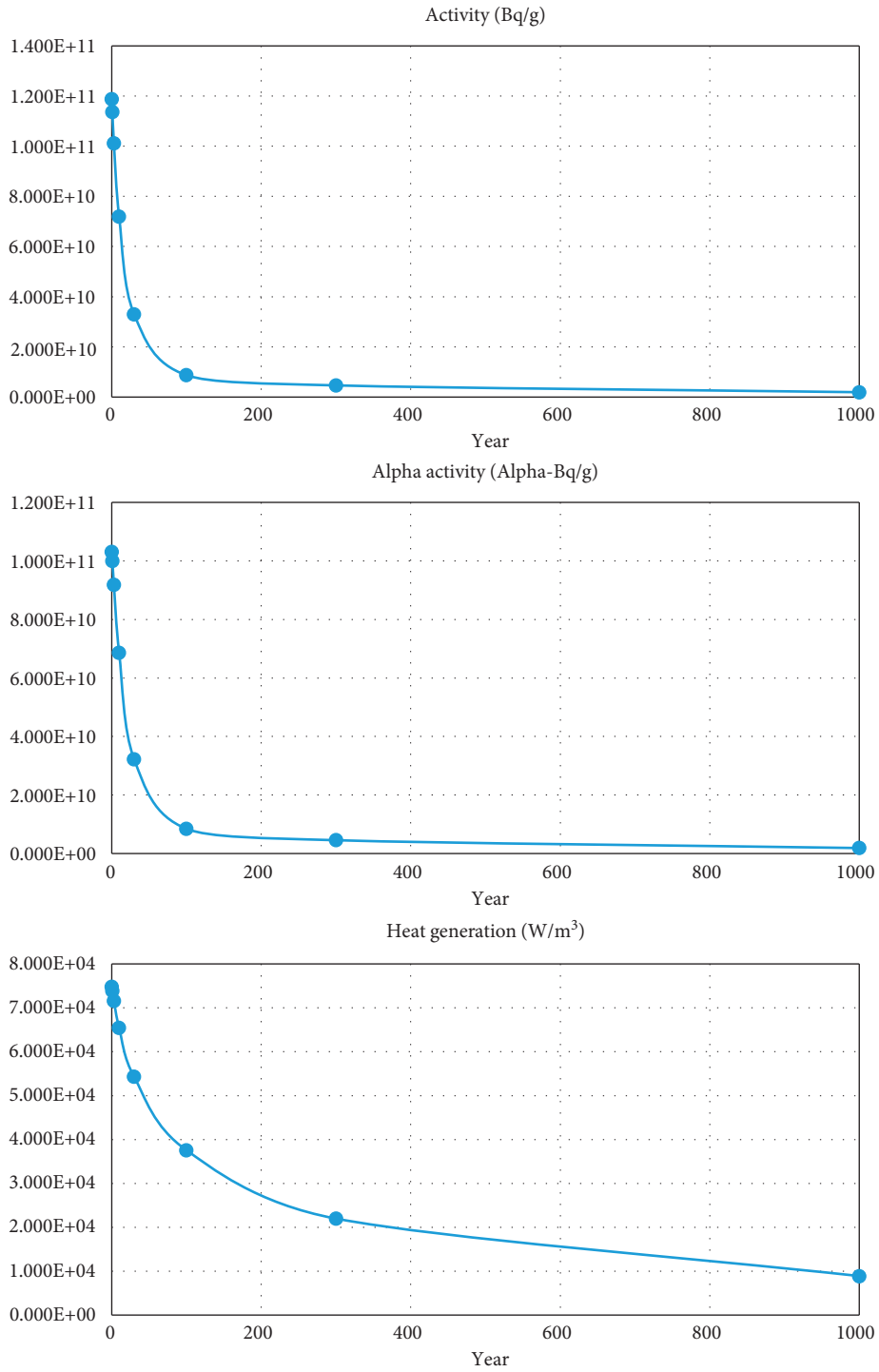


FIGURE 7: Radiological characteristics of TRU/RE waste/waste form.

TABLE 11: Radiological characteristics of Se waste/waste form.

Year	0.3	1	3	10	30	100	300	1000
Bq/g	2.366E+07	2.366E+07	2.366E+07	2.366E+07	2.366E+07	2.366E+07	2.365E+07	2.361E+07
W/m <sup>3</sup>	8.034E-01	8.034E-01	8.034E-01	8.034E-01	8.034E-01	8.032E-01	8.028E-01	8.016E-01

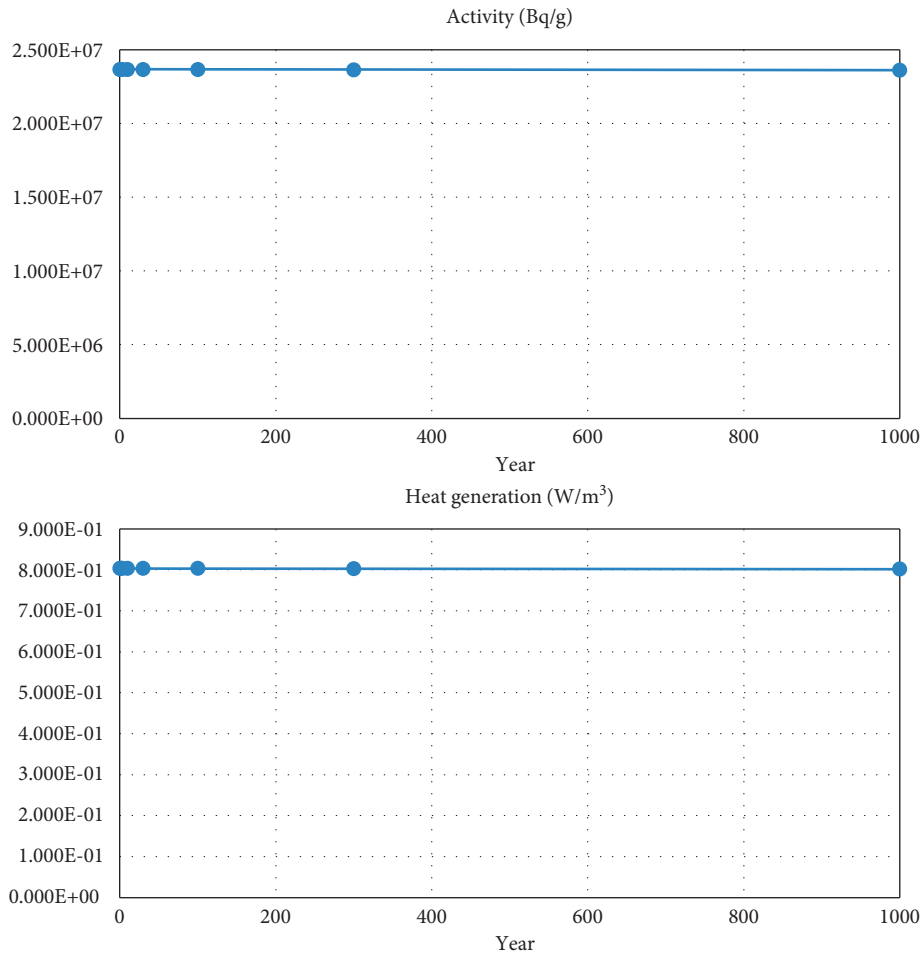


FIGURE 8: Radiological characteristics of Se waste/waste form.

TABLE 12: Radiological characteristics of  $\text{Ca}(\text{ReO}_4)_2$  waste/waste form.

Year	0.3	1	3	10	30	100	300	1000
Bq/g	$3.408E+08$	$3.408E+08$	$3.408E+08$	$3.407E+08$	$3.407E+08$	$3.407E+08$	$3.404E+08$	$3.396E+08$
$\text{W/m}^3$	$2.257E+01$	$2.257E+01$	$2.257E+01$	$2.257E+01$	$2.257E+01$	$2.256E+01$	$2.255E+01$	$2.250E+01$

TABLE 13: Radiological characteristics of  $\text{Ca}_5\text{Re}_2\text{O}_{12}$  waste/waste form.

Year	0.3	1	3	10	30	100	300	1000
Bq/g	$2.109E+08$	$2.109E+08$	$2.109E+08$	$2.108E+08$	$2.108E+08$	$2.108E+08$	$2.106E+08$	$2.102E+08$
$\text{W/m}^3$	$1.515E+01$	$1.515E+01$	$1.515E+01$	$1.515E+01$	$1.515E+01$	$1.515E+01$	$1.514E+01$	$1.510E+01$

nuclide). Owing to the high atomic weight of the Re nuclide compared to that of the Tc nuclide, the characterization was evaluated conservatively using the higher density of the Re-containing waste/waste form. The density of the  $\text{Ca}(\text{ReO}_4)_2$  phase was  $4.885 \text{ g/cm}^3$  and the waste loading was 53.8%. In the case of the  $\text{Ca}_5\text{Re}_2\text{O}_{12}$  phase, the density was  $5.301 \text{ g/cm}^3$  and the waste loading was 33.3%. Based on these values, the radiological characteristics were analyzed, as shown in Tables 12-13 and Figure 9. Tc-99 is a long-lived nuclide with a half-life

of 211,000 years; therefore, it exhibits low initial radioactivity and heat generation. Additionally, it exhibits initial radiological properties of  $\sim 10^8 \text{ Bq/g}$  and  $\sim 10^1 \text{ W/m}^3$ , which are higher than those of Se nuclides ( $\sim 10^7 \text{ Bq/g}$  and  $10^{-1} \text{ W/m}^3$ ). Consequently, the radiological characteristics of the Tc/Se waste/waste forms are dominated by the Tc nuclides. Additionally, owing to the low radioactivity and heat generation of the Tc/Se waste/waste forms, the increase in core temperature in the disposal environment is expected to be negligible.

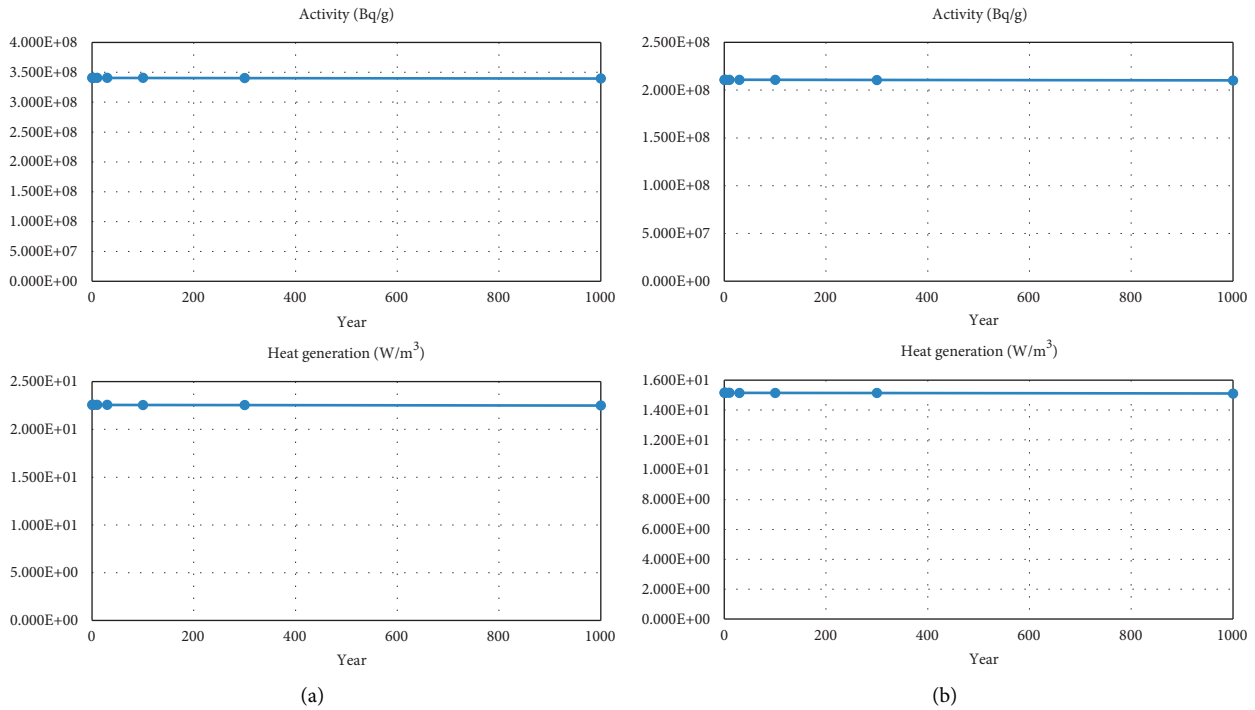


FIGURE 9: Radiological characteristics of Tc waste/waste form. (a)  $\text{Ca}(\text{ReO}_4)_2$  waste/waste form and (b)  $\text{Ca}_5\text{Re}_2\text{O}_{12}$  waste/waste form.

## 4. Conclusion

To reduce the environmental burden caused by the disposal of spent nuclear fuel, the nuclide management process of the waste burden minimization technology develops a nuclide management technology that can maximize the disposal efficiency by selectively separating and collecting major nuclides contained in the spent nuclear fuel. To manufacture a waste form with high durability, a thorough characterization of the waste generated in the process was required. Therefore, in this study, the physical and radiological properties of the waste and waste forms of major nuclides (Cs, Sr, I, TRU/RE, and Tc/Se) were analyzed. Cs and Sr nuclide waste is planned to be manufactured as a waste form in the phase of  $\text{CsAlSi}_2\text{O}_6$  and  $\text{SrTiO}_3$ , which showed high radioactivity and heat generation rates at the initial stage. For TRU/RE nuclide waste, the waste form in the phase of  $(\text{TRU, RE})\text{PO}_4$  was found to be classified as high-level waste, exceeding the high-level waste classification criteria. For I and Tc/Se nuclide wastes, both waste forms (iodapatite/glass waste form for I waste and a mixture of  $\text{CaSeO}_3/\text{Ca}_5\text{Tc}_2\text{O}_{12}/\text{Ca}(\text{TcO}_4)_2$  for Tc/Se waste) showed low radioactivity and heat generation rates owing to their long-lived nuclide. The results of this study could be used to derive the centerline temperature for the thermal stability evaluation of waste forms and for the feasibility evaluation of each disposal system in the development of the waste burden minimization technology.

## Data Availability

The data used to support the findings of this study are included within the article.

## Conflicts of Interest

The authors declare that they have no conflicts of interest.

## Acknowledgments

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