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Volatility and leachability of heavy metals and radionuclides in thermally treated HEPA filter media generated from nuclear facilities

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ABSTRACT

The purpose of the present study was to apply thermal treatments to reduce the volume of HEPA filter media and to investigate the volatility and leachability of heavy metals and radionuclides during thermal treatment. HEPA filter media were transformed to glassy bulk material by thermal treatment at 900 °C for 2 h. The most abundant heavy metal in the HEPA filter media was Zn, followed by Sr, Pb and Cr, and the main radionuclide was Cs-137. The volatility tests showed that the heavy metals and radionuclides in radioactive HEPA filter media were not volatilized during the thermal treatment. PCT tests indicated that the leachability of heavy metals and radionuclides was relatively low compared to those of other glasses. XRD results showed that Zn and Cs reacted with HEPA filter media and were transformed into crystalline willemite ($ZnO{\cdot}SiO_2$) and pollucite ($Cs_2OAl_2O_34SiO_2$), which are not volatile or leachable. The proposed technique for the volume reduction and transformation of radioactive HEPA filter media into glassy bulk material is a simple and energy efficient procedure without additives that can be performed at relatively low temperature compared with conventional vitrification process.

1. Introduction

High efficiency particulate air (HEPA) filters are used to clean air for operations involving a variety of hazardous materials, such as asbestos and radionuclides. HEPA filters are used in medical, military, electronic, and industrial applications, where clean air is essential [1]. Many radioactive HEPA filter wastes are generated from radioactive facilities that are currently in operation, being repaired, or are under decommissioning. Approximately 1500 drums of spent filter waste have been stored in the waste storage facility at the Korea Atomic Energy Research Institute (KAERI) since its initial operation. Among these wastes, approximately 90% is HEPA filter media [2,3]. In the future, HEPA filters with high radioactivity will be created via pyroprocessing, which is a treatment used for spent nuclear fuels.

A HEPA filter consists of a frame, separator, sealant, glass fiber and gasket. The frame and separator are contaminated with low concentrations of nuclides, while HEPA filter media such as glass fiber are contaminated with a high concentration of heavy metals and radionuclides because contaminants, dust, and organic matter were filtered by the media [3]. In addition, the volume of HEPA filter media is large, and the density is low. Therefore, HEPA filter media should be treated by decontamination and volume reduction for safe disposal.

Dissolution is a process wherein filter media are removed from the filter housings and dissolved in an appropriate solvent, such as hydrofluoric acid (HF) and nitric acid (HNO_3). The process dissolves the fiberglass filter media and destroys organic compounds. The dissolution of filter media is a relatively inexpensive process. However, the process produces secondary corrosive liquid waste streams that require further treatment. This secondary waste is difficult to treat due to its high dissolved solid and salt content [4–7].

Incineration is an excellent process for the volume reduction of HEPA filters with wood housings. However, thermal processes for the treatment of waste containing volatile metals present several disadvantages. When the housings of HEPA filters are composed of stainless steel, only the polymer coating on the fiberglass media and seal material are burned [8]. Therefore, the frame must be separated from the HEPA filter to conduct the thermal treatment. Because HEPA filter media are rich in silica (SiO_2), a network glass of former oxides, HEPA filter media can be transformed into glassy materials by thermal treatment [9,10]. Previous studies have shown that vitrification can occur in the absence of additives when the thermal treatment is conducted on solid wastes containing large amounts of SiO_2 [11–13]. Vitrification, the transformation of a liquid into a glassy material, significantly reduces the volume of a material and immobilizes harmful elements [12]. However, studies on the thermal treatment of radioactive HEPA filter media have not yet been conducted.

In the present study, thermal treatment was carried out on HEPA filter media to reduce the volume of the material and to obtain a

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Table 1
The elemental composition (%) of HEPA filter media.

Element	Composition (%)
SiO_2	56.74
B_2O_3	11.29
Na_2O	9.03
Al_2O_3	6.63
BaO	4.48
ZnO	4.44
K_2O	3.62
CaO	3.16
MgO	0.31
Fe_2O_3	0.16
LiO_2 others	0.14
Sum	100

reliable form of waste. The characteristics of the thermal treatment of HEPA filter media were investigated, and vitrification was performed to transform HEPA filter media into glassy bulk material. Radionuclides and heavy metals in radioactive HEPA filter media were analyzed using an X-Ray Fluorescence (XRF) spectrometer for major elements and an Inductively Coupled Plasma-Optical Emission Spectroscopy (ICP-OES) and Atomic Absorption Spectroscopy (AAS) for minor elements, and Multi-Channel-Analysis (MCA) for radionuclides, respectively. In addition, the volatility and leachability of radionuclides and heavy metals in radioactive HEPA filter media were investigated during and after the thermal treatment, respectively. X-Ray Diffraction (XRD) analyses were conducted to investigate crystalline mineral formations associated with immobilized heavy metals and radionuclides at a concentration of 10 wt.% in non-contaminated HEPA filter media.

2. Materials and methods

2.1. Characteristics of the thermal treatment of HEPA filter media

Non-contaminated HEPA filter media produced in Korea Airtech Corporation were analyzed by XRF (RIX 2100, Rigaku, Japan) to investigate the elemental composition. The elemental composition of HEPA filter media is presented in Table 1. The characteristics of the thermal treatment were investigated for non-contaminated HEPA filter media. Thermogravimetric Analysis (TGA) was conducted to determine the characteristics of the thermal treatment as a function of the temperature. A muffle furnace was used in the thermal treatment to investigate the effect of the vitrification temperature (600, 700, 800 and 900 °C). The treatment time was held for 2 h to ensure good compositional homogenization of the melt. XRD (DXR-3000, Rigaku, Japan) analyses were conducted to determine the crystallinity of thermally treated HEPA filter media. These analyses were implemented to select useful conditions for the vitrification of a reliable form of waste.

2.2. Chemical analysis of radioactive HEPA filter media

In total, 5 HEPA filter media samples were collected from the Irradiated Material Examination Facility (IMEF), Radioactive Waste Treatment Facility (RWTF), Post Irradiation Examination Facility (PIEF), Hanaro (HA), and Radioisotope (RI) facility in KAERI. The analytical procedure for the determination of trace metals in filters described in Work Assignment 5-03 [14] was performed to investigate the concentration of heavy metals. The collected filters were extracted in Teflon centrifuge tubes containing 25 mL of 4% nitric acid (Sigma–Aldrich, USA) and were ultrasonicated for 3 h using a heated (69 °C) sonication bath. After sonication, the samples were allowed to cool to room temperature. The samples were filtered using filter paper (541, Whatman, England) and were diluted with distilled (DI) water to obtain a final volume of 50 mL prior to the use of ICP-OES (ICAP6000, thermo elemental, USA) and AAS (5100 model, Perkin Elmer, USA) for the analysis of As, Cd, Co, Cr, Cs, Pb, Se, Sr, U, and Zn. Radioactive HEPA filter media were analyzed using MCA, and a high-purity Ge detector was employed to evaluate the radioactivity of the radionuclides.

2.3. Volatility tests of radioactive HEPA filter media for heavy metals and radionuclides

Radioactive HEPA filter media from the IMEF facility in KAERI were sampled. The frame was separated, and the top (sample 1, S1) and bottom (sample 2, S2) of the HEPA filter media was sampled. The graphite crucible was filled with HEPA filter media. The size of the graphite crucible was 25 cm × 12 cm × 7 cm, and the thickness was 1 cm. The samples were heated in a muffle furnace at 900 °C for 2 h, as described in Section 2.1. The amount of weight loss and volume reduction was measured after the thermal treatment, which was conducted to investigate the volatility of heavy metals (Zn, Pb, Sr, and Cr) and radionuclides (Cs-137 and Co-60) in HEPA filter media, as described in Section 2.2. Samples obtained before and after thermal treatment were digested by acid mixture of HCl-HNO_3-HF (1:1:1) using microwave, similar to that used by Jiri et al. [15] for glass fiber filters and were analyzed by ICP-OES and AAS for heavy metals (Zn, Pb, Sr, and Cr). For the radioactivity analysis, radioactive HEPA filter media were cut into pieces and were added to 20 mL MCA bottles containing quartz (SiO_2). After the thermal treatment, the samples were ground and added into 20 mL MCA bottles containing quartz (SiO_2) to minimize the effect of geometry prior to the thermal treatment. The radioactivity of the material was analyzed using MCA for Cs-137 and Co-60 before and after the thermal treatment.

2.4. Leachability tests of radioactive HEPA filter media for heavy metals and radionuclides

The leachability of individual components was evaluated according to the Product Consistency Test (PCT) leaching method, and the leachant was analyzed by ICP-OES and AAS. The treated samples used in Section 2.3 were hand-pulverized and sieved through −100- to +200-mesh stainless steel sieves to obtain a grain-size fraction of 75–150 μm, which was ultrasonically cleaned with DI water and ethanol and dried in an oven at 90 °C. A 5 g portion of cleaned and thermally treated material was placed into a 150 mL desensitized 304-L stainless steel container, which was filled with 50 mL of DI water, sealed, and stored for 7 days at 90 °C. After 7 days, aliquots of the solution were filtered through a 0.45-μm filter, acidified with 1 vol.% HNO_3, and submitted for ICP-OES and AAS analyses to determine the glass composition (Si, B, Li, and Na), heavy metal content (Zn, Pb, Sr, and Cr), and radionuclide concentration (Cs and Co).

2.5. XRD analyses for thermally treated HEPA filter media

To examine the relevant mineralogy in thermally treated samples, non-contaminated HEPA filter media were prepared with 10 wt.% heavy metals (Zn, Pb, Sr, and Cr) and radionuclides (Cs and Co) to enable the identification of thermally treated samples by XRD. ZnO, PbO, SrO, and CrO_3 were purchased from Sigma–Aldrich (USA), and $Co(NO_3)_2{\cdot}6H_2O$ (Aldrich, USA) and $CsNO_3$ (Sigma–Aldrich, USA) were used as surrogate nuclides. The samples were heated in a muffle furnace at 900 °C for 2 h. The samples were hand-pulverized to pass through a #200-mesh sieve for XRD analyses. Step-scanned XRD patterns were collected by a Rigaku DXR-3000 computer-automated diffractometer (Rigaku, Tokyo, Japan). XRD analyses were conducted at 40 kV and 40 mA using a diffracted beam graphite monochromator and Cu radiation.

The XRD patterns were collected in the 2θ range of 5–100°, and a step size and count time of 0.02° and 3 s per step were employed, respectively.

3. Results and discussion

3.1. Characteristics of melting in non-contaminated HEPA filter media

The elemental composition of *non-contaminated* HEPA filter media was analyzed by XRF. The results indicated that the SiO_2 content was 56.7% and that the B_2O_3, Na_2O, Al_2O_3, BaO, ZnO, K_2O, and CaO contents were 11.3, 9.03, 6.63, 4.48, 4.44, 3.62 and 3.16%, respectively (Table 1). HEPA filter media consists of a glass network of former oxides (SiO_2) and small amounts of modifier oxides (Na_2O, CaO, K_2O, and MgO) and other ceramic oxides (Al_2O_3 and Fe_2O_3), which are responsible for the physico-chemical properties of the final glassy bulk material produced by thermal treatment [10]. The standard base glass used in high-level-waste (HLW) vitrification (termed "MW") in UK is composed of 61.7% SiO_2, 21.9% B_2O_3, 11.1% Na_2O, and 5.4% Li_2O [16,17]. In addition, PAMELA glass produced in Germany–Belgium is composed of 52.7% SiO_2, 13.2% B_2O_3, 5.9% Na_2O, 4.6% CaO, 2.7% Al_2O_3, 18.7% others [18]. The composition of HEPA filter media is similar to that of MW and PAMELA glasses.

TGA analysis of the HEPA filter media was conducted to investigate the characteristics of the HEPA filter media by thermal treatment. The TGA results showed that the HEPA filter shrank at temperatures ranging from 200 °C to 600 °C (Fig. 1), and the corresponding weight loss was equal to 8%. The observed weight loss

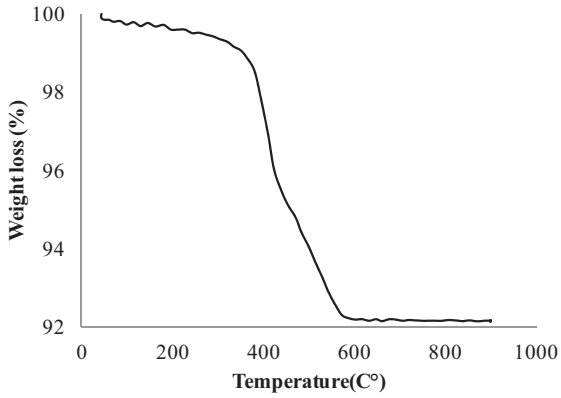

Fig. 1. TGA analysis of HEPA filter media (1 g of non-contaminated HEPA filter media, 10 °C/min, air atmosphere).

was attributed to the dehydration of H_2O, volatilization of combustible binder, and the phase change of the inorganic filter media into a liquid. To investigate the compositional homogenization of the thermal treatment, the proposed procedure was conducted at 600, 700, 800, and 900 °C for 2 h. Volume reduction did not occur at 600 °C, and vitrification (transformation to glassy bulk material) was not observed at 700 °C (Fig. 2). At 800 °C, the volume of HEPA filter media was reduced, but the material was not completely vitrified. In contrast, complete volume reduction and vitrification occurred at 900 °C (Fig. 2). Based on these results, good compositional homogenization of the melt was achieved after thermal

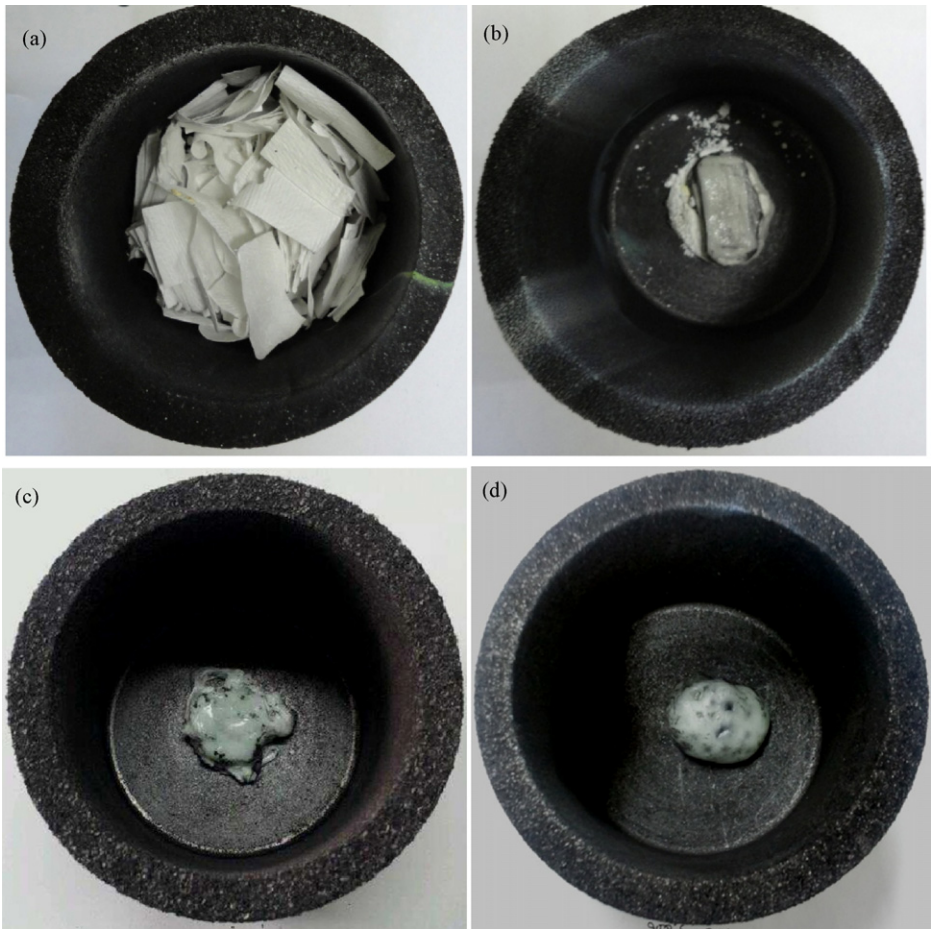

Fig. 2. Non-contaminated HEPA filter media after thermal treatment for 2 h at (a) 600 °C, (b) 700 °C, (c) 800 °C, and (d) 900 °C.

Table 2
The concentration of heavy metals and radionuclides in HEPA filter media generated from IMEF, RWTF, PIEF, HA, and RI in KAERI.

	IMEF	RWTF	PIEF	HA	RI	Average
Heavy metals						
As (μg/mg)	0.130	n.d.	0.028	n.d.	n.d.	0.032
Cd (μg/mg)	n.d.	n.d.	0.013	n.d.	n.d.	0.003
Co (μg/mg)	0.039	n.d.	n.d.	0.029	n.d.	0.014
Cr (μg/mg)	1.183	0.072	0.072	0.064	0.026	0.283
Pb (μg/mg)	2.977	0.870	0.498	0.076	0.050	0.894
Se (μg/mg)	n.d.	n.d.	n.d.	n.d.	n.d.	0.00
Sr (μg/mg)	0.447	0.390	0.245	0.917	0.523	0.504
U (μg/mg)	0.023	0.247	0.020	n.d.	n.d.	0.058
Zn (μg/mg)	15.400	17.667	5.475	26.467	8.300	14.662
Cs (μg/mg)	n.d.	n.d.	n.d.	n.d.	n.d.	0.00
Radionuclides						
Cs-137 (Bq/g)	4050.00	255.00	1.550	0.150	0.210	861.38
Co-60 (Bq/g)	4.150	33.000	n.d.	n.d.	n.d.	7.430
U-238 (Bq/g)	n.d.	n.d.	n.d.	n.d.	n.d.	0.010

treatment at 900 °C for 2 h. XRD analyses were conducted to investigate the crystalline phase of thermally treated HEPA filter media. The XRD analysis indicated that the HEPA filter media was transformed into an amorphous material with a glass structure (Fig. 3).

3.2. Concentration of heavy metals and radionuclides

The concentration of heavy metals and the radioactivity of radionuclides in radioactive HEPA filter media were analyzed. The results indicated that the most abundant heavy metals in the radioactive HEPA filter media was Zn of 14.66 μg/mg, followed by Pb of 0.894 μg/mg, Sr of 0.504 μg/mg, and Cr of 0.283 μg/mg (Table 2). Other metals such as As, U, Se, Co, Cs, and Cd were present at low levels in radioactive HEPA filter media. The Zn concentration in HEPA filter media was relatively high, and raw HEPA filter media contained approximately 4% ZnO. Zn is an important base metal required for various applications in the metallurgical, chemical, and textile industry [19]. The MCA results indicated that the main radionuclide was Cs-137 of 861.39 Bq/g. However, U-238 was not detected. Cobalt is a non-volatile element, and Cs readily volatilizes during waste thermal processes such as vitrification and incineration [20]. A number of radionuclide partitioning studies on waste from thermal plants have shown that Cs is the most troublesome radionuclide due to its high volatility [21]. Therefore, the emission of volatile radioactive Cs species is one of the greatest health risks associated with waste thermal plants [20]. In the present study, the radioactivity of Cs-137 in HEPA filter media was high because volatilized Cs-137 was filtered using HEPA filter media.

3.3. Volatility test for HEPA filter media

Volatility tests were performed for heavy metals and radionuclides in radioactive HEPA filter media to evaluate volatilization during the thermal treatment. The proposed thermal treatment was conducted on radioactive HEPA filter media under the optimal conditions described in Section 3.1. This result indicated that the volume of HEPA filter media decreased significantly due to the thermal treatment (Fig. 4). In addition, thermally treated HEPA filter media were transformed into glassy bulk material, and a stable solid form was obtained (Fig. 4). With respect to the volatility of heavy metals, the results indicated that Zn, Sr, and Cr did not volatilize during the treatment (Fig. 5). However, approximately 20% Pb was volatilized by the thermal treatment. In S1, 25% Cr was volatilized; however, Cr was not volatilized in S2 (Fig. 5). Cs and Co were not detected by ICP-OES and AAS. In the MCA analysis, the initial

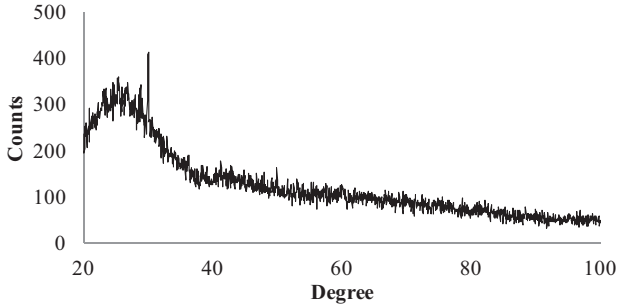

Fig. 3. XRD data for the HEPA filter media thermally treated at 900 °C for 2 h.

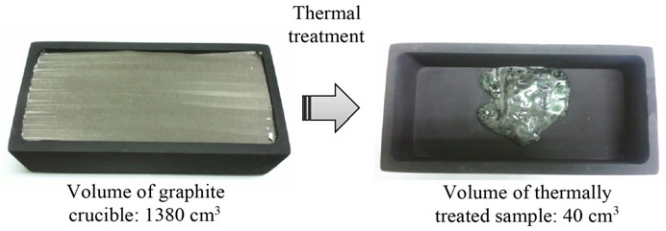

Fig. 4. Radioactive HEPA filter media before and after thermal treatment for 2 h at 900 °C.

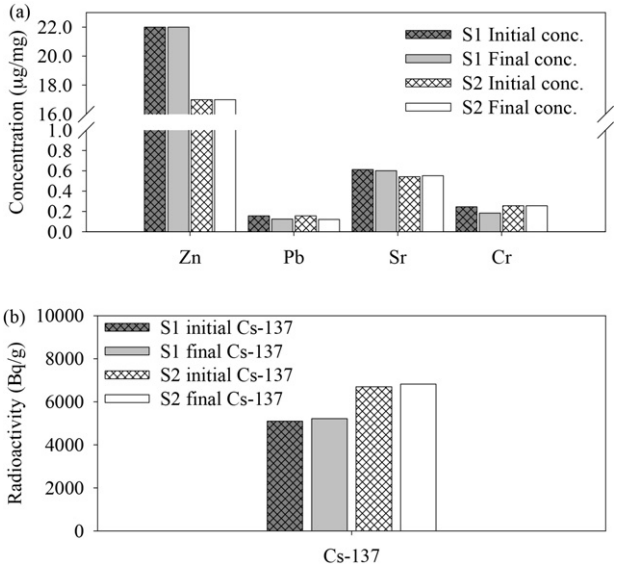

Fig. 5. Concentration of (a) heavy metals and (b) radionuclides in radioactive HEPA filter media before and after thermal treatment. The sample 1 (S1) and sample 2 (S2) of the HEPA filter media was taken from top and bottom in HEPA filter. Experimental conditions −900 °C for 2 h.

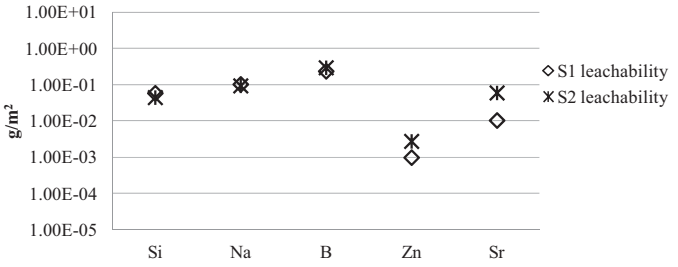

Fig. 6. PCT tests for the leachability of heavy metals and radionuclides in radioactive HEPA filter media after thermal treatment. The sample 1 (S1) and sample 2 (S2) of the HEPA filter media was taken from top and bottom in HEPA filter. Experimental conditions −900 °C for 2 h.

Cs-137 content in S1 and S2 was 5098 and 6697 Bq/g, and the final Cs-137 content in S1 and S2 was 5220 and 6819 Bq/g, respectively (Fig. 5). Cs-137 was not volatilized, and the concentration of Cs-137 increased after the thermal treatment due to the reduction in volume and weight loss, which enriched the material. The radioactivity of initial Co-60 was relatively low. These results indicated that the proposed procedure can be used to treat HEPA filter media, and a capturing system for the volatilization of Zn, Sr, Cr, and Cs-137 is not required.

Yang et al. [20] reported that ZnO and CrO_3 did not volatilize until a temperature of 900 °C was attained under atmospheric conditions. These results are consistent with those of the present study. In contrast, in a previous investigation, a small amount of PbO was volatilized at 900 °C [20]. In the present study, approximately 20% Pb was volatilized at 900 °C. Strontium is a semi-volatile species that volatilizes at temperatures greater than 1000 °C [20]. In the present study, at 900 °C, SrO did not volatilize. Cobalt is a non-volatile species, and Cs is volatile, as mentioned in Section 3.2. In the current study, Co-60 and Cs-137 did not volatilize. In the chemical analysis, Cs was not detected by ICP and AAS, indicating that the Cs concentration was low. Therefore, low concentrations of Cs completely reacted with the HEPA filter media, and volatilization did not occur. Sill [22] demonstrated that Cs vaporizes from a calcined mixture, and complete volatilization is achieved within 1 h at 1300 K. However, after a viscous melt forms, the volatility of Cs decreases markedly. Bibler et al. [23] compared the measured and predicted concentration of technetium-99 (^{99}Tc) and ^{137}Cs in glass produced from Savannah River tank waste and found that 31% of ^{99}Tc and 4% of ^{137}Cs were vaporized during vitrification. Taylor [24] performed a detailed analysis on the volatility of ruthenium and Cs during several vitrification processes and reported that the volatility of ^{137}Cs was approximately 1%; however, values as high as 13% have also been measured. These results indicated that the volatilization of Cs significantly decreases due to vitrification of HEPA filter media.

3.4. Leachability tests of HEPA filter media

PCT tests of heavy metals and radionuclides in radioactive HEPA filter media were conducted to evaluate the stability of the material after thermal treatment. The thermally treated samples used in the volatility tests were evaluated in the leachability tests. Fig. 6 shows the results of PCT leachability tests performed on thermally treated samples. The leach rate (LR) was calculated according to the following equation:

$$LR_i = \frac{m_i}{f_i S t} \quad (1)$$

In Eq. (1), m_i is the amount of element i leached into solution (g), f_i is the weight fraction of element i in the solid, S is the surface area of the test sample (0.0299 m^2/g), and t is the duration of the test (day) [25]. As shown in Fig. 6, the leach rates of Si, Na, B, Zn, and Sr were 4.53×10^{-2}, 9.53×10^{-2}, 3.01×10^{-1}, 2.81×10^{-3}, and 6.09×10^{-2} g/m^2 in S1, and 5.96×10^{-2}, 1.05×10^{-1}, 2.40×10^{-1}, 1.00×10^{-3}, and 1.06×10^{-2} g/m^2 in S2, respectively. The leach rates of Li, Cs, Pb, Cr, and Co were not detected. MCA analysis showed that the radioactivity of the leachant was 1.08 and 3.11 Bq/cm^3 in S1 and S2, respectively. Approximately 0.02 and 0.05% Cs-137 were released from thermally treated samples. The composition of Li in the HEPA filter media was low (0.4 wt.%), and Li was not detected in the PCT tests. To develop glass media and obtain a stable form of waste, the leach rate of major glass components such as Si, B, Li, and Na should be lower than that of the environmental assessment (EA) glass. The leach rate of B, Li, Na, and Si was 8.36, 4.8, 6.67, and 1.96 g/m^2 in EA glass [26]. In the present study, the leach rate of B, Li, Na and Si was lower than that of the EA glass. PCT tests on the Pacific Northwest National Laboratory (PNL) HLW glass indicated that the leach rate of B, Li, Na, and Si was 0.260, 0.333, 0.256, and 0.154 g/m^2, respectively [26]. The leach rate of the thermally treated samples was lower than that of the PNL HLW glass. The leach rate of the glass components (Si, B, Li, and Na) was relatively low compared to the PCT results of other glassy materials, indicating that the samples satisfied the criteria for radioactive waste.

Park et al. [27] studied the stabilization/solidification of radioactive salt waste using SiO_2–Al_2O_3–P_2O_5 (SAP). According to the PCT-A method, the leach rate of Cs and Sr was approximately 1×10^{-3} g/m^2 day (7×10^{-3} g/m^2). In the present study, a leach rate of 1.4×10^{-3} g/m^2 was obtained for Sr, which was lower than the results of Park. However, the leach rate of Cs was not detected by the PCT test. The leach rate of Sr for high sodium waste in borosilicate glasses is 1×10^{-6} to 1×10^{-7} g/cm^2 day (7×10^{-2} to 7×10^{-3} g/m^2), and the leach rate of Cr, Mn, Fe, Co, and Ni is 1×10^{-7} to 1×10^{-8} g/cm^2 day (7×10^{-3} to 7×10^{-4} g/m^2) [28]. The leach rate of heavy metals and radionuclides in HEPA filter media was lower than that of compared to the other PCT results, indicating that the heavy metals and radionuclides remained in a stable form after the reaction with the HEPA filter media. XRD analyses were conducted to identify new crystalline phases for HEPA filter media reacted with heavy metals and radionuclides.

3.5. XRD analyses for heavy metals and radionuclides reacted with HEPA filter media

To obtain more information on thermally treated samples, XRD analyses were performed on non-contaminated HEPA filter media reacted with 10 wt.% heavy metals and surrogate nuclides, and the results are shown in Fig. 7. The HEPA filter media reacted with ZnO, SrO, and $CsNO_3$ and were transformed into willemite ($ZnO{\cdot}SiO_2$), strontium silicate ($SrSiO_3$), and pollucite ($Cs_2OAl_2O_34SiO_2$) after thermal treatment, respectively (Fig. 7a, c, and f). However, new crystalline material containing PbO, CrO and $Co(NO_3)_2{\cdot}6H_2O$ was not identified. The volatility and leachability of Zn and Sr was relatively low, possibly due to the transformation of HEPA filter media reacted with Zn and Sr into willemite ($ZnO{\cdot}SiO_2$) and strontium silicate ($SrSiO_3$), respectively, which are stable forms. HEPA filter media reacted with $CsNO_3$ was transformed into pollucite ($Cs_2OAl_2O_34SiO_2$), which is also a stable form.

To form a willemite (Zn_2SiO_4) phase, conventional methods require calcinations at temperatures greater than 1000 °C and long reaction times, and these processes limit the shape and size of the particles. However, studies on hydrothermal and sol-gel techniques have shown that the crystallization of willemite (Zn_2SiO_4) can be achieved at temperatures of 100 °C [29]. In the present study, willemite was formed by thermal treatment at 900 °C for 2 h. The addition of low concentrations of ZnO to alkali borosilicate (typically <5 wt.% ZnO) confers several beneficial effects, including

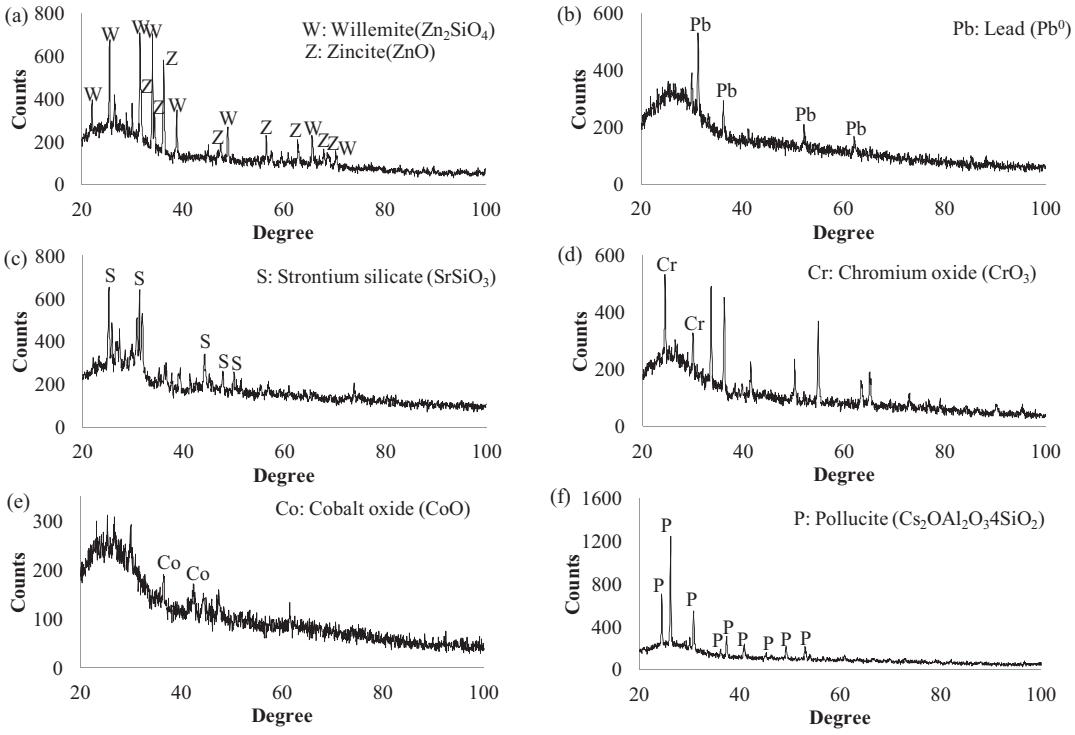

Fig. 7. XRD data for HEPA filter media reacted with (a) ZnO, (b) PbO, (c) SrO, (d) CrO_3, (e) $Co(NO_3)_2{\cdot}6H_2O$, and (f) $CsNO_3$ and thermally treated at 900 °C for 2 h.

improved chemical durability and mechanical processing ability [30–32]. In contrast, ZnO acts as a nucleating agent when present at low concentrations in alkali silicate and aluminosilicate glasses [33].

A secondary Cs phase is often found in altered simulated nuclear waste glasses [34]. Vasil'eva et al. [35] demonstrated that, in the absence of sodium, Cs is immobilized in natural Cs-containing crystalline phases such as pollucite. Research on the aqueous alteration of Japanese simulated waste glass showed that the majority of Cs dissolved from glass exists in the alternation phase due to the presence of a reversible phase or the incorporation of Cs into analcime, pollucite or solid solutions [36]. Curti et al. [37] subjected simulated nuclear waste glass based on pollucite ($Na_{0.3}Cs_{0.7}[H_2O]_{0.3}AlSi_2O_6$) to long-term alterations, and found that the distribution and speciation of Cs was different from that of pollucite. In the present study, Cs reacted with HEPA filter media was transformed to pollucite, a stable form, which indicated that the volatility and leachability was low.

Based on these results, thermal treatment at 900 °C for 2 h did not volatilize heavy metals and radionuclides, and the leachability of individual components was relatively low, indicating that treated samples are safe for disposal because they are transformed into a crystalline phase by reaction with HEPA filter media. The proposed technique for solidifying radioactive HEPA filter media into mineral-like forms is a simple, energy-efficient procedure without additives, and stable forms that are not prone to volatilization or leaching are obtained due to the reaction with HEPA filter media.

4. Conclusions

The characteristics, volatility, and leachability of thermally treated HEPA filter media were evaluated in the present study. The volume of thermally treated HEPA filter media was significantly reduced, and the filter media was transformed into glassy bulk material. The main heavy metals and radionuclides in radioactive HEPA filter media were not volatilized. The leachability test results showed that the heavy metals and radionuclides remained in a stable form because the radionuclides and heavy metals reacted with HEPA filter media (SiO_2) and were transformed into crystalline willemite ($ZnO{\cdot}SiO_2$), strontium silicate ($SrSiO_3$), and pollucite ($Cs_2OAl_2O_34SiO_2$). Based on these results, thermal treatment is one of the most recommended technologies for HEPA filter media treatment.

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