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De-chlorination and solidification of radioactive LiCl waste salt by using SiO_2-Al_2O_3-P_2O_5 (SAP) inorganic composite including B_2O_3 component

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Abstract–SAP (SiO_2-Al_2O_3-P_2O_5) composite has been recently studied in KAERI to deal with the immobilization of radioactive salt waste, one of the most problematic wastes in the pyro-chemical process. Highly unstable salt waste was successfully converted into stable compounds by the dechlorination process with SAPs, and then a durable waste form with a high waste loading was produced when adding glassy materials to dechlorination product. In the present study, U-SAP composite which is SAP bearing glassy component (Boron) was synthesized to remove the adding and mixing steps of glassy materials for a monolithic wasteform. With U-SAPs prepared by a sol-gel process, a series of wasteforms were fabricated to identify a proper reaction condition. Physical and chemical properties of dechlorination products and U-SAP wasteforms were characterized by XRD, DSC, SEM, TGA and PCT-A. A U-SAP wasteform showed suitable properties as a radioactive wasteform such as dense surface morphology, high waste loading, and high durability at the optimized U-SAP/salt ratio 2.

Keywords: Immobilization, SiO_2-Al_2O_3-P_2O_5, Dechlorination, Vitrification, Radioactive Waste

INTRODUCTION

Pyroprocessing consisting of a series of electrolytic processes is of considerable interest in recovering uranium and transuranic elements from a spent nuclear fuel. The spent fuel goes electrolytic reactions in a molten salt, where U and TRUs (Transuranic elements) are recovered as a metallic form and most of fission products are left in a molten salt. In Korea, LiCl used at the electroreduction process and LiCl-KCl used at the electro-refining process are expected to be generated as a radioactive waste containing small amounts of fission products [1-3]. These salt wastes are very problematic due to the physico-chemical properties of the salt, such as high volatility and low compatibility with conventional glass. Up to now, several options have been suggested to deal with the salt waste. First, waste salt can be purified by using Czochralski crystallization method, and total amount of salt waste could be decreased by recycling the purified salt [4]. Phosphate compounds have been suggested to precipitate fission products after dissolving the salt in water, but remaining species which are highly water-soluble alkali such as Cs and Rb are still a problem [5]. Second, the waste salt can be immobilized with adjusted matrix for final disposal. There are two approaches to the immobilization of the salt wastes. One is the immobilization of the salt with incorporation of Cl. A sodalite ($Na_8Al_6Si_6O_{24}Cl_2$)/glass material, one of the Cl-containing minerals, has been widely studied at the Idaho National Laboratory (INL) [6-8]. Metcalfe and Donald suggested a chlorapatite structure ($Ca_{10}(PO_4)_6Cl_2$) to immobilize $CaCl_2$ waste [9]. However, there remain some problems, such as low waste loading (~10 wt%) and relatively low chemical durability due to intrinsic limitations. Another approach is the immobilization of the salt via dechlorination process. The salt can be converted to an oxide-based material or other stable forms by removing the chloride as Cl_2 or HCl by heat-treatment. Wet conversion method using KOH was suggested by Leturcq et al. [10]. A related option is to add $(NH_4)H_2PO_4$ as phosphate source to the waste for the thermal conversion of metal chloride into phosphate glass, but NH_4Cl or HCl is produced as secondary radioactive waste [11]. In our research group, GRSS (gel-route stabilization/solidification) process was established to convert metal chlorides into stable products using Na_2SiO_3-H_3PO_4-$Al(NO_3)_3$-H_2O system [12,13]. This approach indicates that the phosphate compounds have compatibility with borosilicate glass at some stoichiometry. However, there still remained intrinsic problems of aqueous system such as generation of secondary radioactive waste stream (HCl evaporation). On the basis of previous GRSS study, SAP (SiO_2-Al_2O_3-P_2O_5) compound was developed for conversion of metal chlorides into stable products [14,15]. SAP was synthesized by a conventional sol-gel method. SAP is very unique structure consisting of three or four distinctive domains which are sequentially bonded, Si-O-Si-O-Al-O-P-O-P. Remarkable increase of chemical durability and high waste loading for LiCl salt waste up to 40% were achieved by using SAP [14-17]. After the dechlorination reaction, the products can be fabricated as a monolithic wasteform by adding a glassy material.

In this study, another SAP composition was prepared to make a

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[‡] This article is dedicated to Prof. Seong Ihl Woo on the occasion of his retirement from KAIST.

Table 1. Batch composition of U-SAP wasteforms

	Ratio 1	Ratio 1.5	Ratio 2	Ratio 3	Ratio 5	Ratio 9
SiO_2	32.00	35.31	37.23	39.37	41.27	42.64
Al_2O_3	12.40	13.68	14.42	15.25	15.99	16.52
P_2O_5	18.27	20.16	21.26	22.48	23.57	24.35
B_2O_3	7.61	8.39	8.85	9.36	9.81	10.14
Fe_2O_3	1.64	1.81	1.91	2.02	2.12	2.19
Li_2O	22.81	16.78	13.27	9.36	5.88	3.38
BaO	2.65	1.95	1.54	1.09	0.68	0.39
SrO	1.41	1.04	0.82	0.58	0.36	0.21
Cs_2O	1.20	0.89	0.70	0.49	0.31	0.18
	100.00	100.00	100.00	100.00	100.00	100.00
Waste loading (amount of metal chloride)	50.0%	40.0%	33.3%	25%	16.7%	10%

monolithic wasteform without a glassy binder. A new inorganic composite, U-SAP, has boron oxide in the SAP composition. By using U-SAP prepared by a sol-gel process. The U-SAP wasteforms with various waste loadings of salt waste were prepared via dechlorination process. This study reports that the physical and chemical properties for dechlorination products and wasteforms were investigated to fine the optimized composition for a monolithic wasteform without a glassy binder.

EXPERIMENTAL

1. U-SAP and Wasteform Fabrication

Reagent grade source chemicals were chosen. The inorganic composite SiO_2-Al_2O_3-P_2O_5 (SAP) with B_2O_3 and Fe_2O_3, here referred to as Upgraded SAP (U-SAP), was prepared by a sol-gel process. The previous study about the optimization of SAP composition was considered for the composition selection of U-SAP [14]. Tetraethyl orthosilicate (TEOS, adrich 98%), aluminum chlorides ($AlCl_3 \cdot 6H_2O$, Junsei 98%), phosphoric acid (H_3PO_4, Junsei 85%), iron chloride ($FeCl_3$, Aldrich 97%) and boric acid (H_3BO_3, Junsei 99.5%) were used as sources of Si, Al, P, Fe and B, respectively. The molar ratio of Si/Al/P/Fe/B for U-SAP was adjusted to 1/0.456/0.483/0.039/0.412. All reagents were dissolved in EtOH/H_2O and the mixture was under a gel aging process at 70 °C for two days. After the gel aging process, the hydrogel was dried at 110 °C for three days and then heat-treated at 600 °C for 2 h. A simulated waste salt was composed of LiCl, CsCl, $SrCl_2$ and $BaCl_2$ (Aldrich 99% purity) with a composition of 90, 2, 3, and 5 wt%, respectively. Exact composition of actual LiCl salt waste generated from pyroprocessing is still not confined. Major nuclides in LiCl waste are $SrCl_2$ (~7 wt%) and $BaCl_2$ (~15 wt%) and small amounts of RbCl, CsCl, and $EuCl_3$ exist. In this study, CsCl, $SrCl_2$, and $BaCl_2$ were considered as candidate nuclides. The prepared U-SAP was reacted with the simulated waste salt at 650 °C for dechlorination reaction with adjusted U-SAP/waste salt ratios to 1, 1.5, 2, 3, 5 and 9, respectively. Total amount of U-SAP and waste salt for one batch was 45 g. Each batch was labeled ratio-n where the n was assigned to the ratio of U-SAP/waste salt. These ratio-n labels were retained in the figures and tables presented in this study. After the dechlorination reaction, these products were heat-treated at 1,100 °C for 4 h for vitrification process. Targeted compositions of wasteforms are presented in Table 1.

2. Characterization

Dechlorination products and wasteforms were characterized by X-ray powder diffraction (XRD, Rikaku Cu Kα radiation) and Thermogravimetric analysis (TGA, 10 °C/min, 200 ml Ar/min, SEIKO 6300, Japan). Surface images of wasteforms were obtained using a Hitachi S-8010 Field emission scanning electron microscope (FE-SEM) with an accelerating voltage of 10 kV. The samples were sputter-coated with platinum. Physical properties of U-SAP wasteform of ratio 2 such as glass transition temperatures (T_g), thermal conductivity (k), thermal capacity (Cp) and microhardness were analyzed using differential scanning calorimetry (DSC, NETZSCH-DSC 404 C) and Vickers hardness tester. ^{31}P and ^{11}B NMR patterns of wasteforms were recorded using a solid 400 MHz NB NMR spectrometer (Agilent 400 MHz 54 mm NMR DD2). The chemical durability was measured by the PCT-A (Product Consistency Test, PCT) method, according to the procedure in the American Society for Testing and Materials (ASTMs) C1285-02 [18]. About 10 g of each wasteform was crushed and sieved. 20 g of deionized water was added to 2 g of sieved powder in a Teflon vessel, which was maintained at 90 °C for seven days. After the PCT test, leachates were filtered using a 0.45 μm syringe filter. The concentration in leachate was measured by inductively coupled plasma-emission spectroscopy (ICP-ES). The normalized leaching rates of elements, LR_i (g $m^{-2}day^{-1}$), were calculated through the following equation;

$$LR_i = \frac{m_i}{f_i\left(\frac{A}{V}\right)t}$$

where m_i (ppm) is the concentration of the element in a solution, f_i is the mass fraction of the element in the waste form, A/V (2,270 m^{-1}) is the ratio of the surface area to the volume, and t is test duration (day).

RESULTS AND DISCUSSION

1. Dechlorination Reaction Properties Depending on U-SAP/Salt Ratio

XRD results of the dechlorination reaction products with differ-

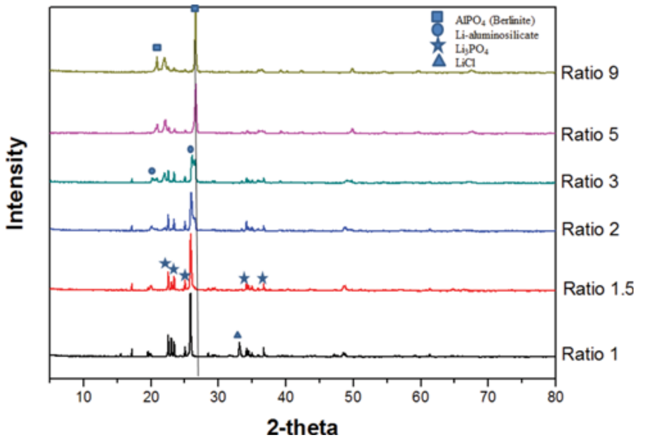

Fig. 1. **XRD results of the dechlorination reaction products with different U-SAP/salt ratio.**

ent U-SAP/salt ratio are shown in Fig. 1. The main crystalline phases were changed depending on increasing U-SAP/salt ratio. Li_3PO_4 and Li-aluminosilicate were major crystalline phases of ratio 1 sample after the dechlorination reaction. The peak of $AlPO_4$ increased gradually as a change of reaction ratio and it was main crystalline phase above ratio 5. In the case of ratio 3, Li_3PO_4 and Li-aluminosilicate coexisted with $AlPO_4$. Reaction products involved with metal chlorides were not detected from XRD analysis. They were maybe below XRD detection limits due to small amounts. In previous study, the dechlorination reaction products for LiCl, CsCl, $SrCl_2$ and $BaCl_2$ with SAP material were composed of metal-aluminosilicate, metal-aluminophosphate and metal phosphate [14,15]. Unstable P-rich region of SAP structure was attacked by LiCl, and a series of active sites such as Si-O-Al, Al-O-P and P-O-P were generated. These active sites were reacted with metal chloride. Chemical reactions for dechlorination reaction with SAP are presented in Eqs. (1)-(4).

$$LiCl+SAP \rightarrow Li_3PO_4+Li_xAl_xSi_{1-x}O_{2-x}+Cl_2 \quad (1)$$

$$LiCl+CsCl+SAP \rightarrow Li_3PO_4+Cs_2AlP_3O_{10} +(Li, Cs)\text{-aluminosilicate}+Cl_2 \quad (2)$$

$$LiCl+SrCl_2+SAP \rightarrow Li_3PO_4+Li_xAl_xSi_{1-x}O_{2-x}+Sr_5(PO_4)_3Cl+Cl_2 \quad (3)$$

$$LiCl+BaCl_2+SAP \rightarrow Li_3PO_4+Li_xAl_xSi_{1-x}O_{2-x}+Ba_5(PO_4)_3Cl+Cl_2 \quad (4)$$

Unreacted LiCl was indicated in ratio 1 sample, meaning lack of reactant, SAP, for dechlorination reaction with LiCl. Mentioned above, $AlPO_4$ generated from the reaction between residual reactive sites was detected from high U-SAP/salt ratio. The existence of $AlPO_4$, therefore, means that more metal chloride can be reacted with U-SAP at a given reaction condition.

Fig. 2 shows the TGA results of the dechlorination reaction products with different U-SAP/salt ratio. Weight loss around 100 °C caused by a water evaporation was not considered. The LiCl, CsCl and $SrCl_2$, unreacted metal chlorides, were vaporized starting at about 600, 800 and 900 °C, respectively. The weight loss values of ratio 1 and 1.5 over 500 °C were 15% and 4.5%, respectively. Residual LiCl was detected from XRD analysis in the case of ratio 1. Unreacted metal chlorides existed under the given condition. Therefore, ratios 1 and 1.5 are not proper ratio of U-SAP and salt for a perfect conversion reaction. For ratios 2, 3, 4 and 5 product, the weight loss over 500 °C was below 1-2 wt%. From these results, it could be inferred that metal chlorides are almost dechlorinated under the given condition. The weight loss of ratios 2, 3, 4 and 5 over 500 °C might be attributed to chlorapatites, which are reaction products between metal chlorides and SAP and unreacted metal chlorides caused by imperfect mixing between U-SAP and salt at a solid state. From the TGA results, U-SAP/salt ratio 2 sample was enough for perfect dechlorination reaction. $AlPO_4$ peak, which indicates the capability for additional dechlorination reaction for metal chloride was not detected at ratio 2 from the XRD analysis of dechlorination reaction products. It could be concluded that the optimized ratio for dechlorination reaction of U-SAP and salt is about 2.

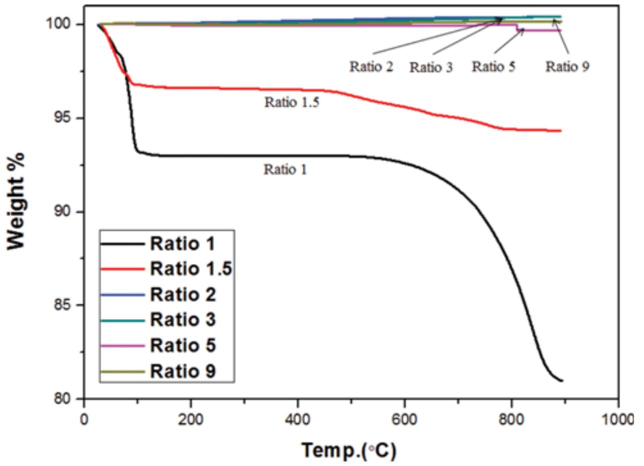

Fig. 2. **TGA results of the dechlorination reaction products with different U-SAP/salt ratio.**

2. Physical and Chemical Properties of Wasteform Depending on U-SAP/Salt Ratio

Bulk shapes of U-SAP consolidated forms heat-treated at 1,100 °C are shown in Fig. 3. Rigid forms were obtained in all samples except ratio 1. The consolidated form of ratio 1 was easily broken when

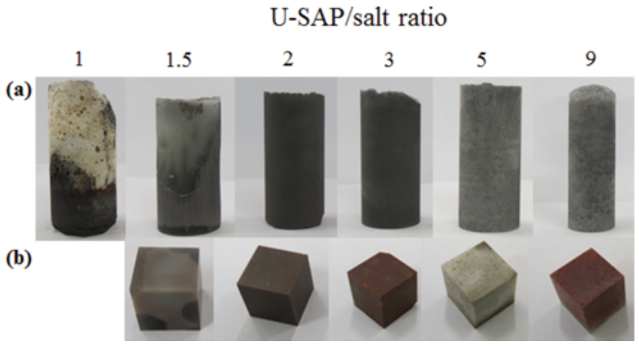

Fig. 3. **Bulk shapes of vitrified wasteforms (a) after just vitrified in cylindrical carbon crucible and (b) after processing to cubic shape.**

making a cubic sample. Cylindrically shaped products presented in Fig. 3(a) were prepared by using a cylindrical carbon crucible, and processed samples to cubic shape by cutting cylindrical forms are shown in Fig 3(b). Bulk separation was detected at ratios 1.5, 3, 5 and 9 samples. Macro pore was formed at ratios 5 and 9. A monolithic form was fabricated only at ratio 2. It is well known that the composition of a network former and modifier in a glass is important to vitrify an inorganic compound. A higher reaction ratio of SAP/salt means lower alkali elements in the product. Based on the results, it could be inferred that ratios 5 and 9 had not sufficient amount of network modifier in the batch composition and they were not largely vitrified, resulting in a product with low density and high fraction of pores. In the previous study, SAP wasteform had two glassy phases, Si-rich phase and P-rich phase, which are not compatible in micrometer scale. At lower U-SAP/salt ratio, considering no peaks of $AlPO_4$ in the reaction product, much of metal chlorides would be converted into phosphate compounds. During the heat-treatment at the given temperature, all the phosphates in the reaction product could not be dissolved into Si-rich or P-rich glassy phase. Also, high amount of phosphate is not compatible to Si-rich phase. Therefore, the excess amount of phosphates or other compounds that was not dissolved in the glassy phase would maintain their own phases even after the consolidation. From these considerations, at the U-SAP/salt 2 in weight has a proper amount of network modifier to a given network former in U-SAP for vitrification, and the proper amount of phosphate formed by the dechlorination reaction would be dissolved into Si-rich or P-rich phase for a monolithic form without bulk separation or macro pores.

Fig. 4 shows XRD results of consolidated forms with different U-SAP/salt ratios. Crystal structures of ratios 1.5, 2 and 3 wasteforms were changed from mixture of Li_3PO_4 and Li-aluminosilicate to generally amorphous phase during the consolidation process. Li_3PO_4 phase partially remained after the consolidation process. SiO_2 and $AlPO_4$ phases were identified from in high U-SAP ratio samples, ratio 5 and ratio 9. It is the reason that unreacted sites of SAP (Si-O-Si, Al-O-P, P-O-P) with metal chlorides are combined with each other under the given temperature condition. Well consolidated forms, including partial crystal phases, were manufactured under a wide range of U-SAP/salt ratio from 1.5 to 3. Waste loading range of metal chlorides calculated from these U-SAP/salt ratios was 25-40%, as shown in Table 1. A wide range of possible reaction ratios for a monolithic form is very essential for the feasibility of U-SAP wasteform in practice due to inaccuracy of waste composition and amount. The properties of developed U-SAP wasteform are comparable to those of other wasteforms. In the previous study, waste of alkali metal chlorides was solidified using sodalite and cancrinite wasteforms with 7.5 wt% waste loading in PNNL [19]. Waste loading of tellurite glass developed for metal chloride immobilization was around 10 wt% [20]. Vance et al. demonstrated up to 10 wt% $CaCl_2$-loadings in chlorapatite ($Ca_{10}(PO_4)_6Cl_2$) structure [21]. The main difference between SAP process and previous studies using sodalite, cancrinite, Te-glass or chlorapatite process is the existence of the dechlorination process. Sodalite, cancrinite, Te-glass or chlorapatite is a Cl bearing wasteform, and waste loading would be limited by its crystalline structure and compatibility with a metal chloride. In this study, U-SAP can convert metal chloride into oxide-based compounds that are very easy to immobilize into a conventional glass. Therefore, the waste loading depends on the composition of network modifier in a target glassy form; alkali element in a waste salt functions as a network modifier in a glass. Based on the results, the U-SAP can treat the waste salt up to about 40 wt% of its own weight and the composition is well fitted with a glass formulation.

Microstructures of the U-SAP wasteforms with different U-SAP/salt ratios were characterized by SEM analysis. In this study, SEM analysis for ratio 1 was not performed because it is meaningless as a wasteform. Fig. 5 shows the SEM images of the U-SAP wasteforms. Dense microstructures without crack were detected in ratios 1.5, 2, and 3, while ratios 5 and 9 had many pores, which would be formed by the entrapment of gas due to the low mobility of glass with low alkali oxides as network modifier. Final compositions of U-SAP wasteforms analyzed by SEM-EDS are tabulated in Table 2. B and Li elements cannot be detected from SEM-EDS and the rest of the compositions except Li and B are normalized to amounts of Si which is not evaporated under our experimental condition. The characterized compositions were well matched to purposed batch compositions. Radioactive elements (Sr, Cs, Ba) were hardly evaporated. In the case of Cl element, it was not detected because most amount of Cl was removed at dechlorination step and remaining Cl after dechlorination was evaporated during vitrification process at 1,100 °C. It had good agreement with TGA results. There is the possibility of existence for trace of chloride compounds as apatite formation such as $Sr_5(PO_4)_3Cl$ and $Ba_5(PO_4)_3Cl$. However, Cl was almost perfectly removed and wasteforms were stable even though small amount of Cl undetected by XRD remained as a formation of apatite. Fig. 6 Shows the NMR patterns of (a) ^{31}P and (b) ^{11}B for ratios 1.5, 2, and 3 wasteforms. For the ^{31}P NMR spectra, a sharp peak at 8 ppm was assigned to the tetrahedral P (PO_4^{-3}) with neighboring Li_3O_4 and a shoulder peak at −30 ppm was assigned to Q^3 P sites connected with -O-P, -O-Al, or -O-Si [14,22]. In the case of ratio 1.5, Q^3 peak was not detected because there was no excess of P over the amount forming Li_3PO_4 to connect with Si, Al or P itself. The ^{11}B NMR patterns were deconvoluted to BO_3 peak at ~11 ppm and BO_4^- peak at ~2 ppm

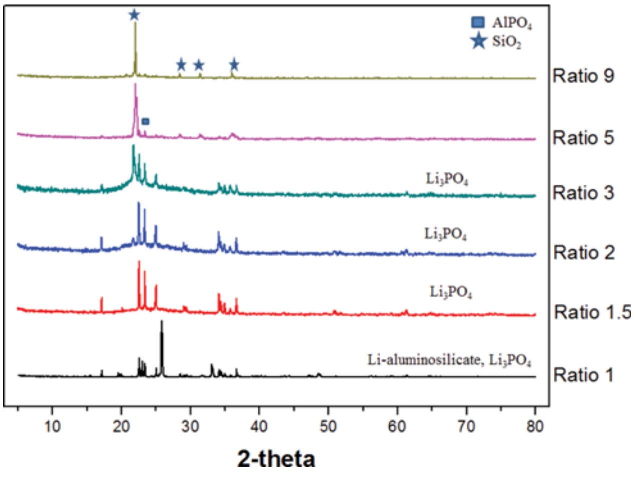

Fig. 4. XRD results of the U-SAP wasteforms.

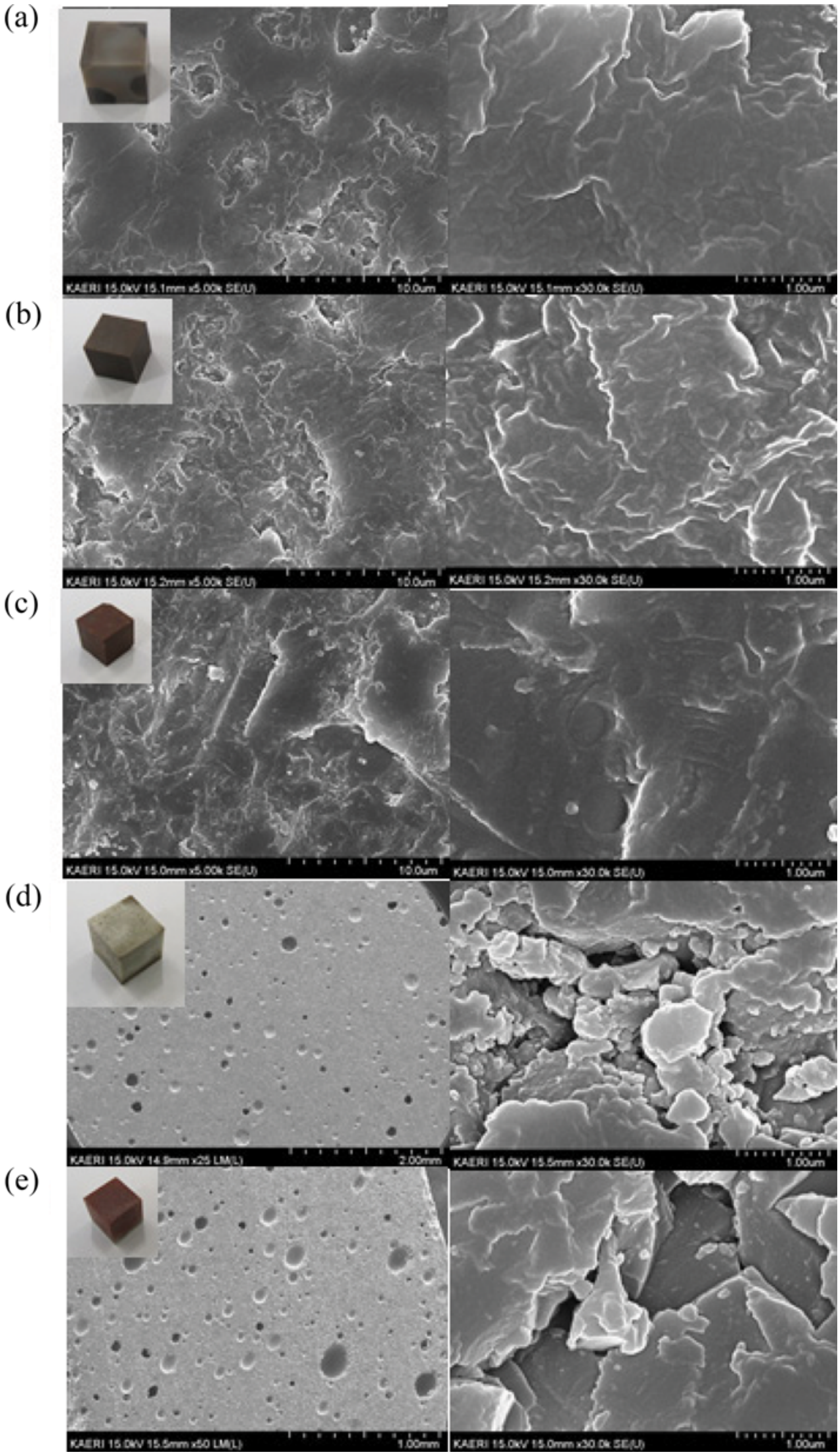

Fig. 5. SEM images of the U-SAP wasteforms.

[23,24]. Characterized wasteforms showed similar BO$_4^-$ to BO$_3$ ratio, which is 1.5. B component in U-SAP wasteforms has two roles in the structure. One is a local charge compensator associated with Li$^+$, Cs$^+$, Sr^{2+}, Ba^{2+} etc. in the case of BO$_4^-$. The other is a modifier which forms non-bridging oxygen atom to connect with other components in the case of BO$_3$. These roles are similar to

Table 2. EDS results of U-SAP wasteforms

	Ratio 1.5	Ratio 2	Ratio 3	Ratio 5
SiO_2	35.31	37.23	39.37	41.27
Al_2O_3	14.5	17.71	18.05	21.05
P_2O_5	19.98	20.06	20.18	24.45
B_2O_3	-	-	-	-
Fe_2O_3	1.45	1.81	1.98	3.02
Li_2O	-	-	-	-
BaO	2.61	2.07	1.51	0.98
SrO	1.51	0.88	0.5	-
Cs_2O	0.81	0.71	0.5	-
Cl	-	-	-	-

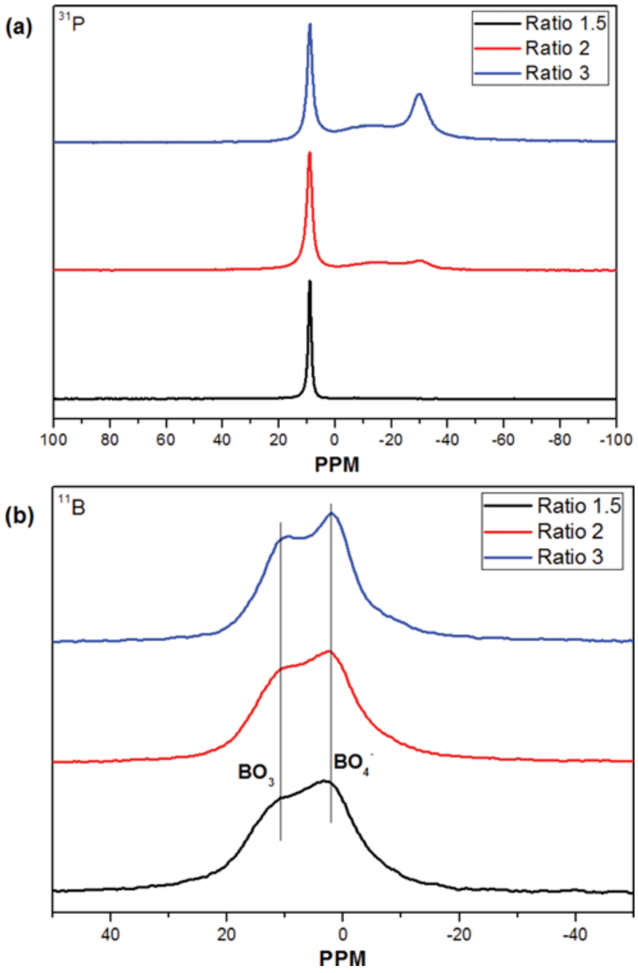

Fig. 6. NMR patterns of Ratio 1.5, 2 and 3 wasteforms (a) ^{31}P and (b) ^{11}B.

roles of B in general borosilicate glasses.

The chemical durability of U-SAP consolidated forms was measured by PCT-A test. Fig. 7 indicates the results of PCT-A leaching test. Normalized leaching rates of matrix components such as Si, Al, P, B, Fe and Li were about 1×10^{-2} g·cm^{-2}day^{-1}, respectively, for U-SAP consolidated forms of ratios 1.5, 2 and 3. Normalized leaching rates of surrogate nuclides, Cs, Sr and Ba, were about 1×10^{-3} g·m^{-2}day^{-1}, respectively, except a wasteform of ratio 1. Especially, the wasteform of ratio 3 shows the lowest leaching rates of Cs (1.23×10^{-3} g·m^{-2}day^{-1}), Sr (1.48×10^{-3} g·m^{-2}day^{-1}) and Ba (2.02×10^{-3} g·m^{-2}day^{-1}). This leach resistance of these nuclides and matrix components is similar to previous SAP study in KAERI [15]. Leaching resistance is not great affected by addition of boron to SAP. U-SAP consolidated forms exhibit very low leaching rates of the nuclides compared with that of the environmental assessment (EA) glass (0.97 g·cm^{-2}day^{-1}) [25]. Leaching rate of low-activity reference material (LRM) glass for Na reported by ANL was 0.07 g·m^{-2}day^{-1} [26]. Riley et al. in PNNL reported similar Na leaching rate of tellurite glass system (0.07 g·m^{-2}day^{-1}) with that of LRM glass [20]. In the leach resistance, the U-SAP wasteform is comparable to other wasteforms.

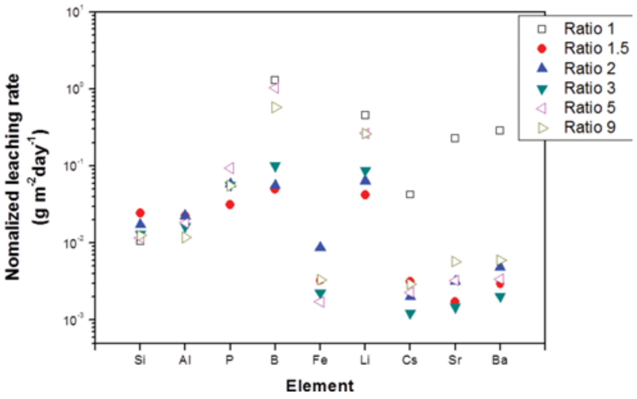

Fig. 7. Normalized leach rate of the U-SAP wasteforms tested by PCT-A (75-150 μm powdered samples, 90 °C for 7 days).

Table 3. Physical properties of U-SAP wasteform with Ratio 2

Physical properties of U-SAP wasteform with Ratio 2	
Density (ρ)	2.36 g/cm^3
Thermal conductivity (k)	1.1 W/Mk
Thermal capacity (Cp)	0.91 J/Gk
Glass transition temperature (Tg)	480±3 °C
Microhardness	490±15.9 Hv

Physical properties of the U-SAP wasteform with ratio 2 were analyzed by Archimedes principle, DSC and Vickers hardness test as a radioactive wasteform. Measured properties are listed in Table 3. The properties have similar values compared with silicate and phosphate glassy wasteforms [27]. In the case of borosilicate wasteforms, density, Tg, thermal conductivity and microhardness generally were ~2.6 g·cm^{-3}, ~500 °C, ~1.1 W·m^{-1}K^{-1} and ~700 Hv, respectively. Density, Tg, and thermal conductivity of Pb-Fe-phosphate were ~4.7 g·cm^{-3}, ~477 °C and ~0.6 W·m^{-1}K^{-1}, respectively. U-SAP wasteform showed very similar physical properties to the conventional glassy wasteforms.

CONCLUSIONS

Surrogate radioactive metal chlorides wastes composed of LiCl, CsCl, $SrCl_2$ and $BaCl_2$ were stabilized and solidified by U-SAP composite via dechlorination reaction process. U-SAP composite was synthesized by using a sol-gel method. Boron was added to SAP composition to remove additional input of glass material. U-SAP/salt ratio 2 showed the optimized dechlorination reaction ratio, where almost metal chlorides go dechlorination reaction. $AlPO_4$ can be considered as the reaction indicator for the efficient reaction ratio. At proper condition, the U-SAP wasteform showed a monolithic form without crack or pore. The result of PCT-A test showed that the U-SAP has high leach-resistance and it was comparable to other wasteform. In conclusion, by adding boron to the original SAP, a monolithic wasteform could be obtained without a glass binder, and the physio-chemical properties of U-SAP wasteform showed similar to those of other conventional wasteform.

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