Vitrification of Radioactive Waste Salt Using Modified Si-Al-P Composites

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1. Introduction

SAP (SiO2-Al2O3-P2O5) composite has been suggested by KAERI to immobilize the radioactive waste salts generated from pyroprocessing. In Korea, LiCl used at electro-reduction process and LiCl-KCl used at the electro-refining process are generated as radioactive waste salts containing small amounts of fission products. Vitrification method using conventional glass matrix are not suitable to deal with these reactive salts wastes. However, SAP composite successfully convert the highly unstable salt waste to stable compounds via dechlorination process. SAP waste forms exhibit good performances such as high waste loading, high chemical durability and suitable physical properties. In previous study, SAP composites were synthesized by sol-gel method. This method is expensive and is not suitable for mass production to treat large amounts of salt wastes. Therefore, development of economical method to treat salt wastes without performance loss is needed.

2. Experimental

Same composition of previous SAP material was selected. Adjusted amounts of SiO_2, $Al(OH)_3$, $(NH_4)H_2PO_4$, B_2O_3, Fe_2O_3 were mixed. The mixture was heat-treated at 500℃ and the material containing $AlPO_4$ or SAPO-20 was synthesized. LiCl with 0wt%, 10wt% 20wt% and 30wt% ratio of nuclides ($SrCl_2$, CsCl) were used as surrogate radioactive salt waste. The prepared salt waste and K-SAP were mixed with various ratio for wasteform synthesis. Each sample was labeled K-SAP n where the n was assigned to the ratio of K-SAP/waste salt. Additionally amount of nuclides was indicated at end of labels ; K-SAP2-1 (10wt%), K-SAP2-2 (20wt%), K-SAP2-3 (30wt%). Dechlorination process of the mixture was heat-treated at 650℃ and dechlorination product was vitrified with 1300℃ heat-treatment. XRD, SEM, and TGA were conducted for physical characterization. Chemical durability was evaluated using ASTM Standard C 1285-02 (PCT-A).

3. Results and discussion

From XRD result of K-SAP, $AlPO_4$ and Fe_2O_3 peaks were indicated. However, $AlPO_4$ peaks were similar with SAPO-20 material. Further analysis is needed. From XRD results of dechlorination products, unreacted LiCl and Lithium chloride borate existed in K-SAP1. Main crystaline phases were Li-aluminosilicate and $AlPO_4$. In higher ratio of K-SAP/salt, unreacted Cl compounds were removed, and Li_3PO_4 and $AlPO_4$ were formed. It is means that over capacity which can react with waste salt remain in K-SAP. Dechlorination ratio was calculated via TGA results. Weight loss value of K-SAP 1 and K-SAP 1.5 was 15wt % and 4.5wt %, respectively deu to unreacted salt compound. For over ratio 2, weight loss over 500℃ was below 1wt%. it means that metal chlorides are almost dechlorinated under the given condition. Therefore, optimized ratio of K-SAP and salt might be ratio 2 with perfect dechlorination reaction. Using ratio 2 of K-SAP, K-SAP 2-1, K-SAP 2-2, and K-SAP 2-3 with surrogate salt was prepared. Uniform wasteforms were formed over 1300℃. Amorphous phases were indicated from XRD results. From SEM images, dense and defect-free surfaces o K-SAP wasteforms were detected. In the case of durability test, PCT-A method was used. Cs and Sr show high durability of 10^{-2} ~ 10^{-3} g $m^{-2}day^{-1}$ and compositions of matrix such as Si, Al, P, B, and Fe show 10^{-1} ~ 10^{-2} g $m^{-2}day^{-1}$.

4. Conclusion

All wasteforms with wide range of composition except K-SAP 1 showed glassy monolithic shapes. A wide range of wasteform ratio for a monolithic form is essential property as wasteform due to inaccuracy of waste compositions and amounts. Using XRD and SEM analysis, amorphous, dense, and monolith wasteforms were detected.

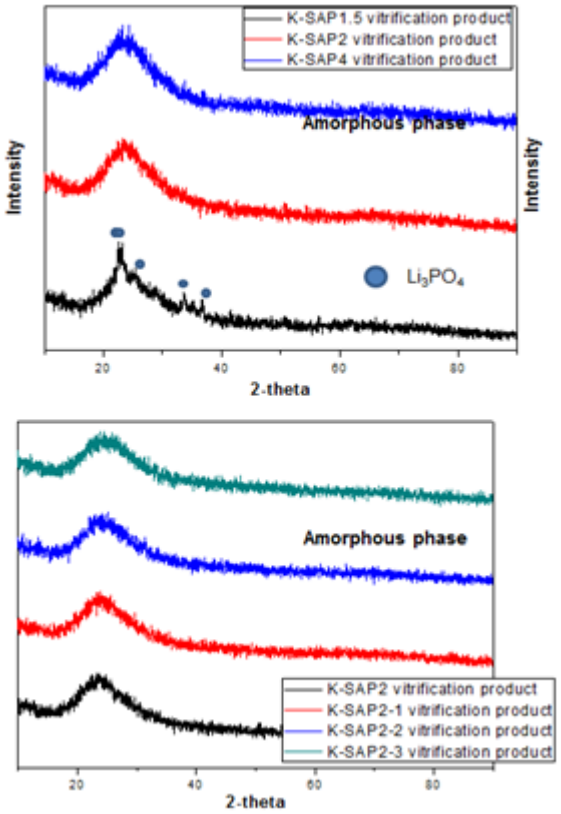

Fig. 1. XRD results of K-SPA wasteforms.

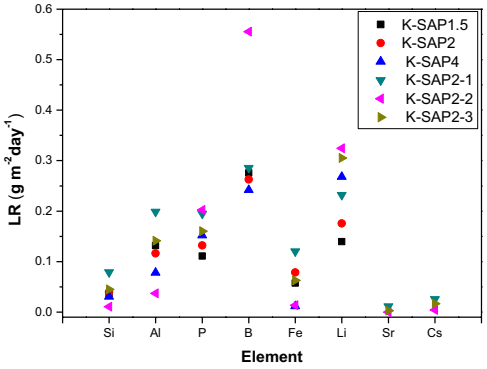

Fig. 2. PCT-A results of K-SAP wasteforms.

REFERENCES

[1] H-S. Park, I-H. Cho, H-C. Eun, I-T. Kim, Y-Z. Cho, and H-S. Lee, "Stabilization/Solidification of Radioactive LiCl-KCl Waste Salt by Using SiO_2-Al_2O_3-P_2O_5 (SAP) Inorganic Composite: Part 1. Dechlorination Behavior of LiCl-KCl and Characteristics of Colsolidation", Environ. Sci. Technol., 45, 1932 (2011).

[2] I.-H. Cho, H.-S. Park, S.-N. Ahn, I.-T. Kim and Y.-Z. Cho, "Stabilization/Solidification of Radioactive LiCl-KCl Waste Salt by Using SiO_2-Al_2O_3-P_2O_5 (SAP) Inorganic Composite: Part 1. The Effect fo SAP Composition on Stabilization/Solidification", J. Kor. Rad. Waste Soc., 10(1), 45 (2012).