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Granulation of Dried Borate Concentrates from Nuclear Power Plants

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1. Introduction

Borate concentrated wastes generated from liquid waste evaporator of 12 pressurized water reactors (PWRs) have been processed to be dry using concentrated waste drying system (CWDS) in Korea. Borate wastes had been stabilized or solidified using paraffin as a binder since 1995, but the solidification process is stopped and concentrated wastes are just stored in drums and on standby at present because paraffin waste forms have some sorts of problems such as heterogeneous forming (stratification), low compressive strength, and high leaching rate [1]. According to No. 2005-18 of Notices of the Ministry of Science and Technology (MOST), all of low- and intermediate level waste (LILW) forms must satisfy the requirement of solidification before they are delivered to the final repository in Kyeongju [2].

In this study, polymer solidification technology is proposed to treat borate concentrated wastes. Originally, this technology has been developed and used to solidify the spent ion exchange resin in many countries. Most commercial polymer binders include epoxy, polyester and vinylester styrene, etc. Recently in May 2003, Idaho National Engineering and Environmental Laboratory (INEEL) issued a report confirming that APS™ (Diversified Technologies Services), a kind of polymer waste form, met the NRC's Waste Form requirements for Class B and C wastes. The Conference of Radiation Control Program Directors (CRCPD) reviewed the INEEL report, and the E-5 Committee issued a letter of waste form approval for the APS™ process. This serves as a national approval in the US, replacing the now-defunct NRC Topical Report Program.

Granulation of dried powder waste is prerequisite for the successful operation of polymer solidification process. Granular wastes permit the maximum waste loading, in situ process without in-line mixing or in-drum mixing, easy workability without dispersion of contaminants, and possible connections with CWDS. A feasibility study on the granulation of dried borate wastes using liquid sodium silicate as a granulating agent was performed.

2. Methods and Results

Through reviewing many materials, we found that liquid sodium silicate was suitable for a granulating agent of boric acid (BA) powder. It is an inorganic binder and its molecular formula is generally expressed as $Na_2O\text{-}nSiO_2\text{-}xH_2O$. Its chemical structures and properties depend on mole ratio of Na_2O/SiO_2. When an acidic component is added to liquid sodium silicate, pH of sodium silicate decreases by neutralization reaction and viscosity increases by siloxane bond and gelation starts to occur. Generally, increasing rate of viscosity and reaction rate of gelation depends on type of acid, additional volume of acid, concentration of solution, and temperature, etc:

$$Na_2O\cdot nSiO_2 + H_2SO_4 + (m\text{-}1)H_2O \rightarrow SiO_2\cdot mH_2O + Na_2SO_4$$

And if liquid sodium silicate reacts with metal ions such as Ca, Mg, Al, and Ba, etc., then insoluble metal hydrate silicate, metal hydroxide silicate, and silicic acid are formed simultaneously along with gelation as follows:

$$Na_2O\cdot nSiO_2 + Ca(OH)_2 + mH_2O \rightarrow CaO\cdot nSiO_2\cdot mH_2O + 2NaOH$$

Specifications of liquid sodium silicate used in this study are as listed in Table 1.

Table 1. Properties of liquid sodium silicate

pH	specific gravity (20°C)	Na_2O (%)	SiO_2 (%)	Fe_2O_3 (%)	mole ratio	viscosity (cps, 20°C)
12 ~ 13	> 1.38	9~10	28~30	<0.03	3.12~ 3.40	>200

Figure 1 shows preliminary equipment of granulation in this study. BA powder was first added to mixing vessel and then upper and lower motor operated simultaneously to mix BA powder homogeneously. Liquid sodium silicate was injected to mixing vessel and its mass rate was controlled by pressure regulator. The gelation and granulation occurs slowly as liquid sodium silicate is mixed with BA powder. Different kinds of granules were prepared according to mixing ratio of BA and liquid sodium silicate. It was found that the distribution of granules depends on rotating speed of two motors and mixing ratio of liquid sodium silicate (Figure 2). The optimum loading of liquid sodium silicate was determined by considering workability, homogeneity of granular size, and compressive strength of specimen. Figure 3 shows a photo of scanning electron microscope (SEM) for the surface of solid granular form. It was observed that BA powder was securely encapsulated in honeycomb structure of the mixture.

3. Conclusion

It was demonstrated that liquid sodium silicate successfully worked for a granulating agent of boric acid powder and the final granular products were having a dense and hard structures. The developed granulation process will be directly applied to in situ polymer solidification in the future.

REFERENCES

[1] J.Y. Kim, A Study on Leaching Characteristics of Radioactive Paraffin Waste Forms Generated from Korean Nuclear Power Plants, Ph.D. dissertation, 2002.
[2] Ministry of Science and Technology, Regulation on the Transfer of Low- and Intermediate Level Waste, No. 2005-18 of Notices of MOST, 2005.

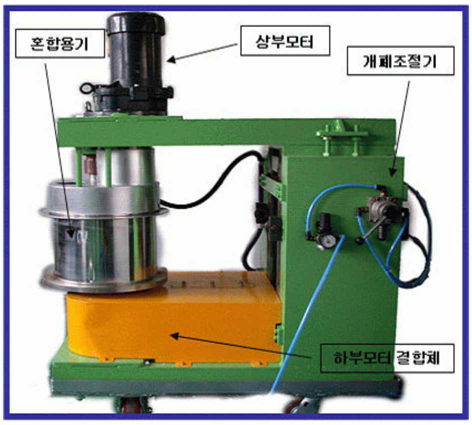

Figure 1. Test equipment of granulation

Figure 2. Granular forms of simulated wastes

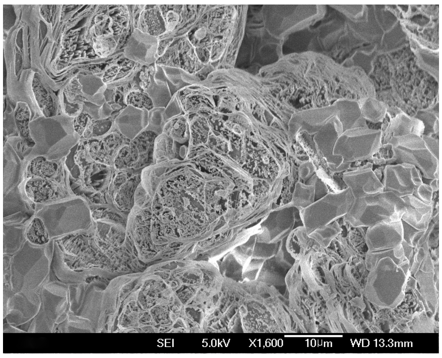

Figure 3. SEM of a granule