Fabrication of Phosphate Cement with High Integrity

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1. Introduction

As the development of industrial society has accelerated, hazardous wastes are generated as well. According to the 1986 statistics of U.S.A, each person made 40 tons of waste in America that year[1]. Treatment of radioactive waste is one of the most important and serious problems related to waste treatments, because its radioactivity and decaying heat have harmful effects to human and environment for a long time. Nuclear developed countries have used conventional method of treatment such as vitrification or cementation in order to stabilize and solidify radioactive waste. Although the former guarantees the formation of high leaching resistant and durable waste form, it requires several hundred (or even more than one thousand) temperature to melt glass frit. This process generates secondary waste volatilized, as well as being non-economical. Cement technology played a role of immobilizing low and middle class wastes. It has advantages of low temperature setting, low cost, easy process, etc. The alkalinity of ordinary cement, however, constrains the utility of cement to the solidification of alkaline waste. In addition, leachability and mechanical strength of cements are not quite appropriate for the stabilization of high level waste. In this regard, chemically bonded phosphate cement(CBPC), which sets by an acid-base reaction, is a potentially expectable material for immobilization of radioactive waste. CBPC not only sets at room temperature, but also encapsulates various isotopes chemically. The performance of CBPC can be enhanced by the addition of fly ash, sand, wollastonite, etc[2]. This study aims at fabricating the CBPC containing fly ash with high integrity. Morphology, microstructure, and compressive strength are evaluated using SEM, and digital compressing machine.

2. Experimental

The process of making CBPC, based on an acid-base reaction, is quite simple: the metal oxide powders are mixed with phosphoric $acid(H_2PO_4)$ or phosphate solution. Commonly used metal oxides are zinc, calcium, magnesium, iron, etc. In this study, we prepared magnesium oxide(Martin Martietta Specialties, USA) and potassium dihydrogen phosphate(KH₂PO₄, JUNSEI) as raw materials. In addition, fly ash(produced at Boryeong) was used as an additive for enhancement of strength. The chemical compositions of fly ash are presented in Table 1. The magnesium oxide powder

used in this experiment was dead burned, which means the powder was calcined at above 1500 $^{\circ}$ C in order to reduce the reactivity by the decrease of specific surface area. If uncalcined MgO is used, the process becomes uncontrollable because of too fast speed of reaction. The basic equation related to this experiment is as follows: MgO + KH_2PO_4 + $5H_2O$ = MgKPO₄ · $6H_2O$. First, two powder materials are mixed to tap water with the above molar ratio. Then, the slurry was stirred during about 30 minutes when magnesia dissolved enough so that reaction occurred. The slurry was poured into cylindrical plastic mould with 5 cm diameter and 10 cm height. The mould was then cured in the forced convection oven at 60 °C. Fly ash added cement was also made through the same procedure of the binder with varying quantity of fly ash. The analyses of microstructure and morphology on cured cement were evaluated using a scanning electron microscope(Philips). The compressive strength of cements was tested using compressor.

3. Results and discussion

Fig. 1 and 2 shows the fractured surfaces of phosphate cements with fly ash of 60 wt% and without fly ash, respectively. It can be seen that that many small spheres, which are cenospheres contained in fly ash, are incorporated in the main glassy phase in figure 1. Small round shaped particles are unreacted magnesia. Rod-like particles shown in Fig. 2 are reaction products. From the morphology of cement containing fly ash, it is suggested that new reaction between fly ash and magnesia or phosphate did not occur. It is noted that, however, fly ash particles were glued together with main amorphous phase, which would enhance the mechanical properties of cement. This inference can be supported by the result of compressive strength test. Table 2 shows the compressive strength data of phosphate cements. As fly ash loading increases, compressive strength also increases. This may be explained as following reasons. First, fly ash fills the void space in cement structure. As the SEM image confirms, fly ash plays the role of filler which makes the cement denser. Second, the cement containing fly ash has low porosity. Because of much heat generation during the reaction, there must be many internal pores in the phosphate cement. If fly ash is added in the cement paste, the heat generation becomes less due to dilution effect, which results in less defect formation in cement.

3. Conclusions

The fabrication and characterization of phosphate cements containing fly ash were carried out. As analyses of microstructure and compressive strength reveal, the properties of phosphate cement can be enhanced by the proper incorporation of fly ash. This result is quite encouraging because the cost of fly ash is relatively low and it is used as a raw material of cesium trapping filter in the head-end process of pyroprocess.

Table I: Chemical Compositions of Fly Ash

Element	Al	Ca	Fe	Κ	Si	С	Na	Mg	Ti
wt%	9.8	7	4.2	1.3	24	4.1	0.7	1.1	0.5



Fig. 1. Morphology of cement containing fly ash of 60 wt%.



Fig. 2. Morphology of cement without fly ash.

Table II: Compressive Strength Data of Fly Ash Containing Cements

Loading(wt%)	30	50	60
Compressive	725	1594	2102
Strength(psi)			

REFERENCES

[1] Environmental Protection Agency, EPA/530 SW-88-011 (1988).

[2] A. S. Wagh, Chemically Bonded Phosphate Ceramics, 21st Century Materials with Diverse Applications, Elevier, pp. 37-41, 2004.