Study on the Dissolution of Concrete for Volume Reduction of Radioactive Concrete Waste

Iksoo Kim*, Maengkyo Oh, and Keunyoung Lee
Korea Atomic Energy Research Institute, 111, Daedeok-daero 989beon-gil, Yuseong-gu, Daejeon, Republic of Korea
*niskim@kaeri.re.kr

1. Introduction

Concrete waste accounts for about 80% of the various types of waste generated from the dismantling of nuclear facilities [1]. Considering the stringent classification criteria and the high disposal cost of radioactive waste in Korea, volume reduction treatment is inevitably necessary to reduce the amount of radioactive concrete waste. The purpose of this research is to develop a technology for the volume reduction of radioactive concrete wastes. As the first step, this research aims to develop a process to dissolve concrete waste and a separation process of dissolved radionuclides. For this purpose, the physico-chemical properties of cement, mortar, and concrete waste powder were investigated, and the dissolution characteristics of concrete powder by nitric acid and hydrochloric acid solution were examined. The optimum conditions for the dissolution of concrete waste powder and radioactive concrete samples contaminated with radioactive isotopes were determined.

2. Experimental

Concrete waste was obtained from a local construction company. This <4 mm pulverized concrete powder was then sieved into 6 sizes (+200 mesh ~ -16 mesh) for use throughout the experiment. For the preparation of radioactive contaminated concrete, standard solutions of radioisotopes (^{60}Co, ^{137}Cs, and ^{90}Sr) were diluted and aliquots of these radioisotopic solutions were injected in concrete powder to have 300 Bq/g radioactivity. These samples were dried for 50 days at room temperature.

After measuring the radioactivity of contaminated concrete powder by using MCA and beta counter, concrete powder was dissolved by adding hydrochloric acid or nitric acid solution of the desired concentration. Undissolved concrete powder were filtered and dried. The weight and radioactivity of undissolved powder was measured again. The residual radioactivity of concrete powder was calculated from the measured radioactivity before and after dissolution.

3. Results and discussion

3.1 Dissolution characteristics of cement based materials

Concrete is a composite material which is made up of filler and binder. The binder (cement paste) glues the filler together to form a synthetic conglomerate. The constituents used for the binder are cement and water, while the filler can be fine or coarse aggregate. Concrete is reacted with acid because of its alkaline nature. Most pronounced is the dissolution of calcium hydroxide which occurs according to the following reaction:

$$2\,HX + Ca(OH)_2 \rightarrow CaX_2 + 2\,H_2O \text{ (HX is the acid)}$$

The decomposition of the concrete depends on the porosity of the cement paste, on the concentration of the acid, on the solubility of the acid calcium salts (CaX_2), and on the fluid transport through the concrete. Acids such as nitric acid and hydrochloric acid are very reactive with concrete as their calcium salts are readily soluble. On the other hand, sulphuric acid is relatively unreactive because of low solubility of its calcium salt. Therefore, in this study, dissolution of concrete was carried out by using nitric acid and hydrochloric acid.

3.2 Dissolution of concrete powder

In order to investigate the dissolution behavior of concrete waste in acidic solution, concrete powder, cement, and mortar were dissolved in 2 M nitric acid solution for 2 hours. About 30wt% of the concrete was dissolved, but more than 93wt% of the cement and 90wt% of the mortar were dissolved. Fig. 1 shows the composition of the dissolved components analyzed by ICP after dissolving the cement, mortar, and concrete in 2 M nitric acid. As shown in figure,

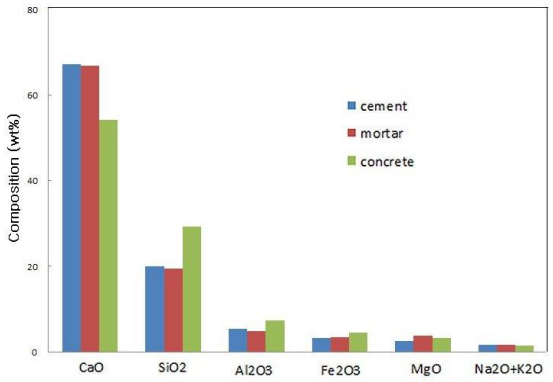

Fig. 1. Chemical composition of dissolved fraction of cement, mortar, and concrete.

the composition of the dissolved solution of cement and mortar is almost the same, and also the composition of the dissolved solution of concrete is similar to those of cement and mortar. In particular, the composition of each chemical component closely coincides with that of Ordinary Portland Cement (OPC). The dissolution ratio of concrete powder in nitric acid solution is about 30wt%, and about 70wt% of the undissolved materials are mostly sand and coarse aggregate. The composition of the dissolved material is almost the same as that of OPC.

The effects of acid concentration and temperature on the dissolution rate of concrete waste powder were examined using hydrochloric acid and nitric acid to determine the optimal dissolution conditions. The weight loss ratio of concrete powder is about 30% on average, which is almost constant even when the acid concentration increases. When the acid concentration is 0.5 M or less, the weight loss ratio increases in proportion to the acid concentration. This indicates that acid is the limiting reactant in the reaction with cement components such as CaO, Al_2O_3, and Fe_2O_3. It was calculated that 0.5 mole of acid was required to completely react with 1 kg of concrete powder.

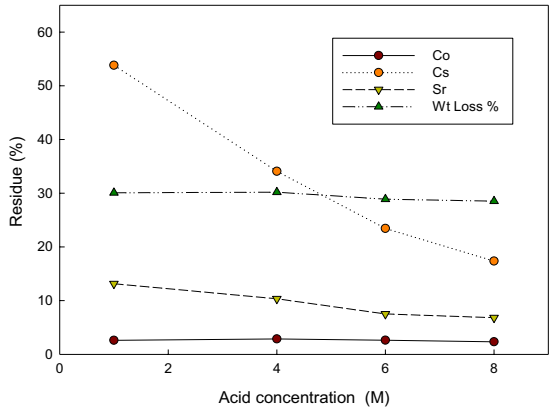

Fig. 2. Decontamination ratio of radioisotopes.

3.3 Dissolution of contaminated concrete powder

Fig. 2 shows the weight loss ratio and the radioactivity residue of the undissolved powders remaining from nitric acid dissolution of the radioactively contaminated concrete powder. Contaminated concrete powder also shows a weight reduction rate of about 30% regardless of the concentration of nitric acid solution when the concentration of nitric acid solution is 0.5 M or higher. The residual ratio of cobalt is constant regardless of the nitric acid concentration, but those of cesium and strontium decrease gradually as the nitric acid concentration increases. However, the radioactive residual ratio of cesium and strontium is higher than that of cobalt even at nitric acid concentration of 8 M.

Dissolution was carried out by increasing the dissolution temperature when the contaminated concrete powder was dissolved again in order to reduce the radioactive residual ratio. Dissolving the primary undissolved powders in 4 M nitric acid solution again for 1 hour, the radioactive residue ratio decreased drastically as the dissolution temperature increased. At 100℃, the radioactivities of cobalt, cesium, and strontium were reduced to less than 3% as compared with the first undissolved powder. The weight loss ratio of the undissolved concrete powder was about 4wt% as compared with that of the first undissolved powder. The total radioactivity retention by primary and secondary dissolution was less than 1% at 100℃ as compared with that of the initial contaminated concrete powder.

ACKNOWLEDGEMENT

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[1] K. Y. Lee et al., “Trends in Technology Development for the Treatment of Radioactive Concrete Waste”, JNFCWT, 16(1), 93-105 (2018).