

VOLUME REDUCTION OF DISMANTLED CONCRETE WASTES GENERATED FROM KRR-2 AND UCP

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As part of a fundamental study on the volume reduction of contaminated concrete wastes, the separation characteristics of the aggregates and the distribution of the radioactivity in the aggregates were investigated. Radioisotope ^{60}Co was artificially used as a model contaminant for non-radioactive crushed concrete waste. Volume reduction for radioactively contaminated dismantled concrete wastes was carried out using activated heavy weight concrete taken from the Korea Research Reactor 2 (KRR-2) and light weight concrete from the Uranium Conversion Plant (UCP). The results showed that most of the ^{60}Co nuclide was easily separated from the contaminated dismantled concrete waste and was concentrated mainly in the porous fine cement paste. The heating temperature was found to be one of the effective parameters in the removal of the radionuclide from concrete waste. The volume reduction rate achieved was above 80% for the KRR-2 concrete wastes and above 75% for the UCP concrete wastes by thermal and mechanical treatment.

KEYWORDS : Volume Reduction, Contaminated Concrete Waste, Aggregate Separation, Radioactivity Distribution

1. INTRODUCTION

Since the decommissioning of nuclear plants and facilities, large quantities of slightly contaminated concrete waste have been generated. In Korea, the decontamination and decommissioning of the retired KRR-2 and UCP at the Korea Atomic Energy Research Institute has been under way. Hundreds of tons of concrete wastes are expected from the Decontamination and Decommissioning (D&D) of these facilities. Several decontamination techniques have been developed to support this maintenance work in nuclear installations. The existing decontamination methods are found to have been destructive, such as pneumatic drilling, while other methods require long term treatment, such as biological methods [1]. Some methods are limited to the material surface, such as chemical methods [2]. The Department of Energy (DOE) has developed concrete decontamination using another strippable coating [3], electro-hydraulic scabbling [4], laser ablation [5-7], dry ice pellet blasting [8], microwave heating [9-14], and electrokinetic [15-19]. The mechanical method has included washing, swabbing, foaming agents, latex-peelable coating, and wet or dry abrasive blasting, as well as grinding of surface and removal of concrete by spalling or scarifying. These techniques are most applicable to the decontamination of a structural surface and are not

appropriate for complicated surfaces where uniform access may not be guaranteed. The existing mechanical technologies have mostly focused on the physical removal of contaminated concrete surface layers.

The proposed treatment technology in this test, which is the integrated thermal and mechanical method, is promising because deeply contaminated materials could be treated and have been selected for the unconditional release of the building and reduction of radioactive concrete waste. The integrated thermal and mechanical technology may provide an essential step for the release of dismantled concrete waste with some complex geometry. Accordingly, thermal and mechanical processes simplify the procedures for monitoring radioactive concrete wastes characterization. Consequently, aggregate materials with reasonably low-activity concentrations may be stored for release in the foreseeable future.

Concrete is a structural material that generally consists of a binder (cement), water, and aggregate. The binder is typically comprised of four principal clinker phase such as tricalcium silicate (Ca_3SiO_5), dicalcium silicate (Ca_2SiO_4), tricalcium aluminate ($\text{Ca}_3\text{Al}_2\text{O}_6$), and calcium aluminoferrite ($\text{Ca}_4\text{Al}_2\text{Fe}_2\text{O}_{10}$). Cement powder (anhydrous cement) created from the co-grinding of clinkers and gypsum is mixed with water and a hydrate phase is formed. The main source of cohesion in cement paste is the nano-particles

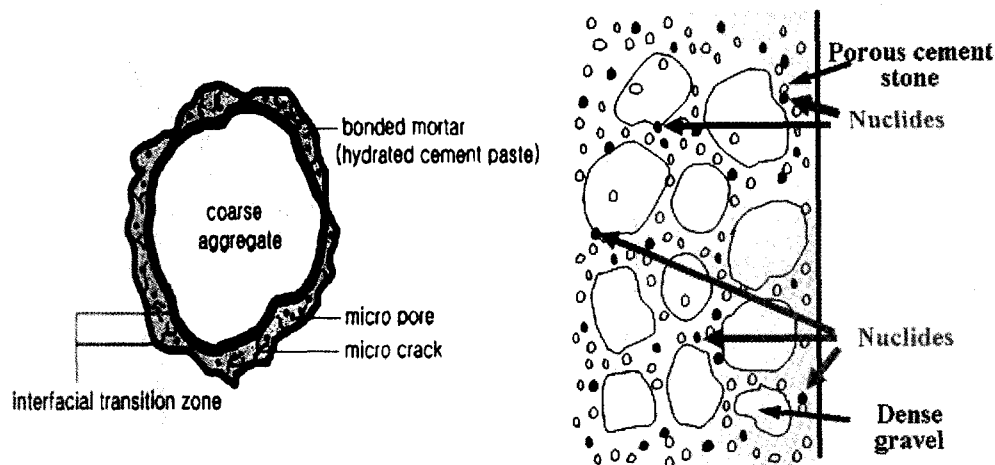


Fig. 1. Scheme of Dismantled Concrete Wastes

of calcium silicate hydrate (C-S-H), which are formed via a dissolution of the original tricalcium silicate (C_3S). The interaction between highly charged C-S-H particles in the presence of divalent calcium counter ions is strongly attractive because of the ion-ion correlations and a negligible entropic repulsion. In the temperature range of 100-300°C, these evolutions mainly have been attributed to the loss of the bound water from the C-S-H gel. Similar consequences have been reported for mortars and concretes [20-23], which have been enhanced sometimes by the appearance of micro-cracks related to the strain incompatibilities between the aggregates (which expand upon heating) and the cement paste (which shrinks upon heating).

Figure 1 shows the scheme of concrete waste [24,25]. The nature of the binding forces in a cement matrix is very species sensitive, so it is difficult to produce a general description. The concrete waste is a basic microstructure form, consisting of C-S-H needles and C-S-H leaves, as well as platy calcium hydroxide and ettringite crystals that grow in a longitudinal shape. Due to the growing crystals, the gap between the cement particles is increasingly bridged. Concrete aggregates are combined strongly with the hydrated cement, including radionuclides. Surface activity can be a loose contamination arising from a deposition of nuclides from an interfacing medium, and it also can be tightly bound. This usually arises from the adsorption of deposited nuclides from cement paste. A mechanical crushing process may provide an essential step for the release of components with complex geometries. Accordingly, a crushing process simplifies the procedures for monitoring radioactive concrete wastes characterization. Consequently, gravel and sand aggregates materials with reasonably low-activity concentrations may be stored for release in the foreseeable future. Potential applications for the limited reuse of the material in the nuclear industry exist, including the fabrication of concrete for certain radiological protection shields, as well as waste storage

containers, backfill or encapsulation materials for waste drums and containers. A recycling or a volume reduction of the concrete wastes through the application of appropriate treatment technologies have merits from the view point of an increase in resource recycling, as well as a decrease in the amount of waste to be disposed of, which results in reduced disposal costs and an enhanced disposal safety.

The aim of this study was to separate clean aggregates as gravel and sand formed contaminated dismantling concrete wastes that were produced from KRR-2 and UCP by thermal and mechanical processes.

2. EXPERIMENTAL PROCEDURE

The ^{60}Co radioisotope was artificially used as a model contaminant for non-radioactive crushed heavy weight concrete taken from the KRR-2 and UCP. The separation was performed by using the thermal method in an electric furnace and mechanical method in a ball mill. The chemical compositions of the concrete waste are summarized in Table 1.

Figure 2 shows the experimental procedure for the separation of radionuclide from the dismantled concrete wastes. The concrete wastes were crushed for a size reduction. In the jaw crusher, the input concrete was crushed to parts with a diameter of about 40 mm. The crushed non-radioactive concrete waste sample was submerged for 24 hours in a diluted radioisotope solution that consisted mainly of ^{60}Co and was dried for 30 days at room temperature. The initial specific radioactivity of the light and heavy weight concrete sample was 28 Bq/g and, 21 Bq/g for ^{60}Co , respectively. The initial specific activity of the sample was measured with a Multi-Channel-Analysis (MCA) of a high-purity germanium (HPGe) detector. The contaminated concrete waste was heated at 300, 500, and 700°C in a furnace for 1 hour. The heated

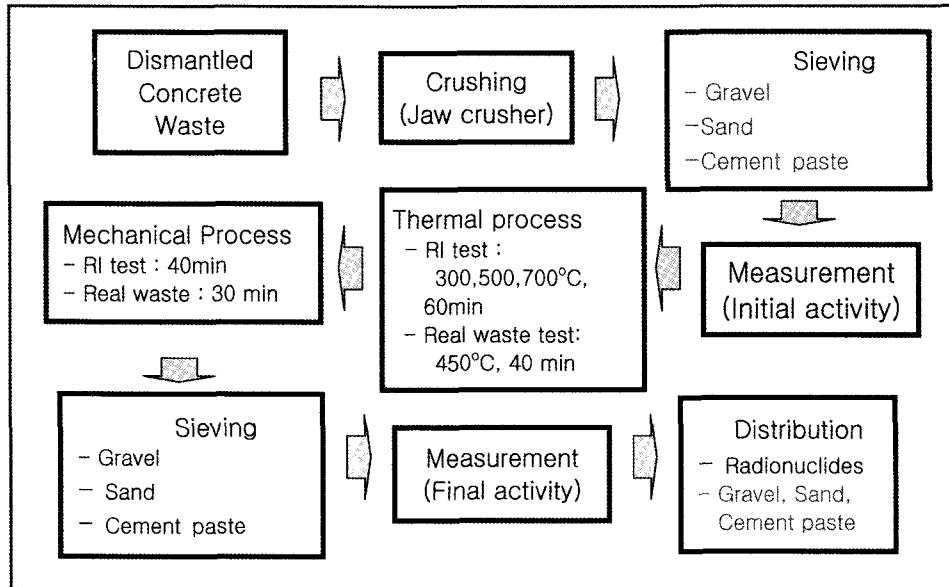


Fig. 2. Experimental Procedure

Table 1. Chemical Composition of the Dismantled Concrete Wastes

Composition	Light concrete (wt%)	Heavy Concrete (wt%)
Fe ₂ O ₃	3.6	39.6
CaO	18.5	16.8
Al ₂ O ₃	9.2	3.3
MgO	1.7	3.9
SiO ₂	61.4	29.3
Na ₂ O	1.3	0.4
K ₂	2.0	1.3
S	0.2	0.2
Etc.	2.1	5.7

concrete waste was crushed by a mechanical method and separated into gravel aggregate, sand aggregate, and cement paste by sieving. The classified aggregate was taken by a standard cylindrical vial at 20ml for activity analysis. The specific activity of aggregates were measured with a HPGe detector to evaluate the radionuclides distribution. To evaluate the distribution of cobalt and uranium, activated heavy weight concrete taken from the KRR-2 site and light weight concrete from the UCP site were used. The dismantled concrete wastes were crushed for a size reduction by the jaw crusher. The separated concrete rubble was heated to about 450°C in an electric furnace for 40 min. The crushed concrete rubble was separated into groups of gravel aggregate (>5mm), sand aggregate (1-5mm), and

cement paste (<1mm). The classified aggregates were analyzed with a MCA.

3. RESULTS AND DISCUSSION

3.1 Contaminated Concrete Waste with Radioisotope

The changes in the specific radioactivity of the separated aggregates from the light and the heavy weight concrete wastes according to heating temperature are given in Figures 3 and 4. As shown in these figures, the specific activity of the separated aggregate decreased by increasing the heating temperature. After thermal and mechanical treatment, the specific activity of the gravel aggregates on light weight concrete decreased by 1.35 Bq/g, 0.33 Bq/g, and 0.44 Bq/g at 300°C, 500°C, and 700 °C. The sand aggregate decreased by 3.9 Bq/g, 2.4 Bq/g, and 2.3 Bq/g. The specific activity of the heavy aggregate after the second mechanical treatment was similar to light weight concrete. The specific activity of the heavy gravel aggregate measured with a HPGe detector was 1.95 Bq/g at 300°C, 0.75 Bq/g at 500°C, and 0.58 Bq/g at 700°C. The contamination level of the sand was higher than the gravel. The contamination level of the gravel and sand aggregates remarkably decreased after thermal and mechanical treatment. It is well known that most of the radioactivity exists in cement mortar and paste composed of concrete [24,25]. The volume reduction of concrete waste was based on separating radioactive concrete into clean aggregates and a radioactive cement paste, which can be readily performed by thermal process in order to weaken the adherence force between the cement matrix

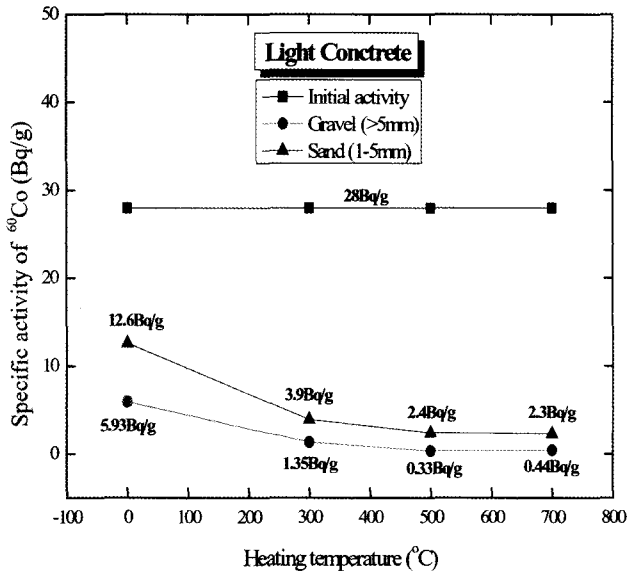


Fig. 3. Change of the Specific Activity on the Light Weight Concrete Aggregates by Heating Temperature

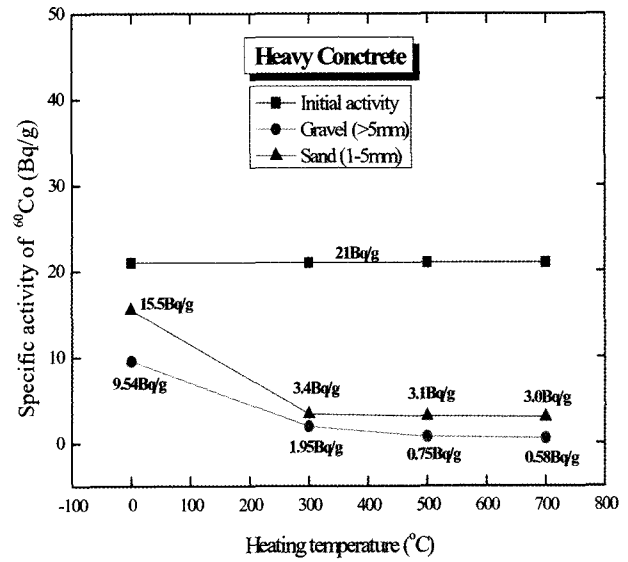


Fig. 4. Change of the Specific Activity on the Heavy Weight Concrete Aggregates by Heating Temperature

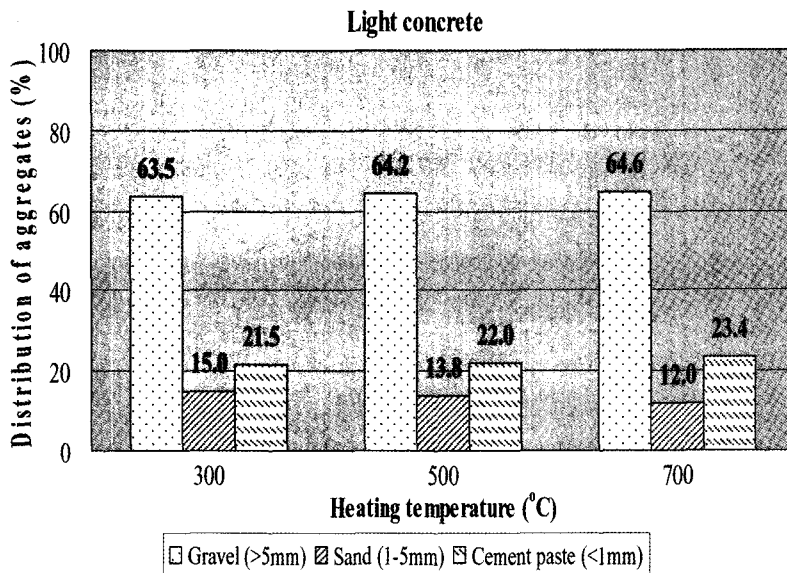


Fig. 5. Distribution of Aggregates on the Light Weight Concrete Wastes by Heating Temperature

and the aggregates, followed by mechanical processes.

Generally, when the concrete waste is heated to approximately 300°C, the cement paste becomes brittle due to dehydration. It has been reported that the thermal and mechanical treatment is quite effective in the removal of the radionuclides from concrete wastes [25]. Bonen and Sarkar [26] have reported ettringite accommodating Co²⁺ at the M²⁺ site in a crystal structure, which Nishita et al. showed was strongly sorbed onto the CSH gel [27]. Vespa et al. [28] found that for samples prepared in air, Co (II) was oxidized to Co (III) after 1hr of hydration

time and that the relative amount of Co (III) increased with hydration time. Co (II) was predominately present as a Co-hydroxide-like phase and/or Co-phyllsilicates, whereas Co (III) tended to be incorporated into a CoOOH-like phase and/or Co-phyllomanganates. Samples prepared in the absence of oxygen revealed solely the presence of Co (II), indicating that oxygen plays an important role in Co oxidation in cement.

Figures 5 and 6 show the distribution of aggregates on concrete waste for a thermal process, followed by a mechanical process. The volume reduction of light weight

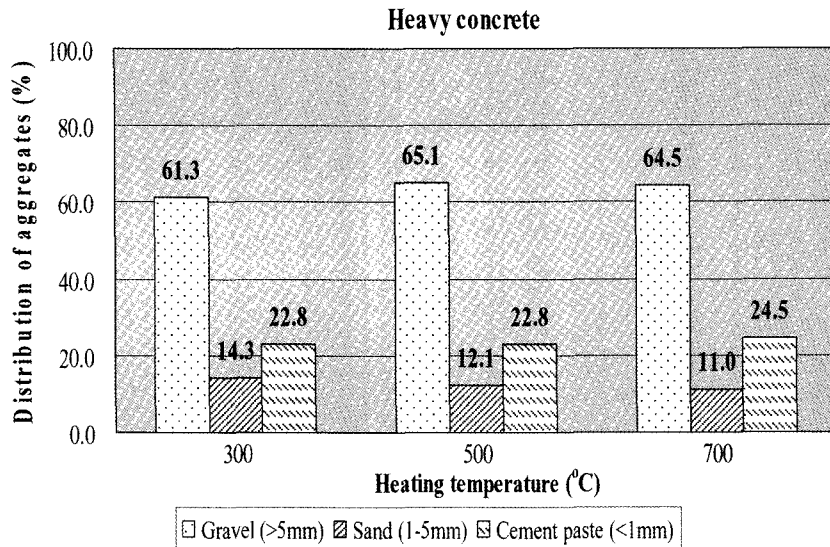


Fig. 6. Distribution of Aggregates on the Heavy Weight Concrete Wastes by Heating Temperature

concrete waste was achieved by up to 65% at 500°C and 700°C with a low specific radioactivity. As shown previously in Figure 3, the specific activity of gravel aggregate on the light weight concrete was detected by 0.44 Bq/g at 700°C. For the heavy weight concrete waste, about 65% can be reduced by the thermal and mechanical process at 700°C. The quantity of the recovered aggregate was found to be closely related to changes in the heating temperature, which was found to have been effective in play an important role in the removal of cement paste from concrete waste.

3.2 Activated Heavy Weight Concrete Waste

The heavy weight concrete used in parts of the nuclear installations was comprised of additional high-density aggregates, especially barite and iron oxides (magnetite, hematite). In situ production of radionuclide in activated heavy weight concrete can occur when it is exposed to a high neutron flux, e.g. in the concrete shielding of reactors (biological shield) and other neutron sources, as well as accelerators. Since it is independent of the primary contamination pathway, radionuclide may then be remobilized in aqueous or other solutions to some degree. Thus, the surface contamination of concrete can be redistributed with time into the concrete matrix by diffusion through a continuous system of water-filled pores and, especially, in cracks. The major nuclides of the gamma emitter were ⁶⁰Co, ¹³⁴Cs, ¹⁵²Eu, and ¹⁵⁴Eu in an activated heavy weight concrete. The activated heavy weight concrete waste used in this experiment only detected ⁶⁰Co. The aggregates were then analyzed only for ⁶⁰Co radionuclide distribution.

Figure 7 shows the distribution of the aggregates for the activated heavy weight concrete by thermal and

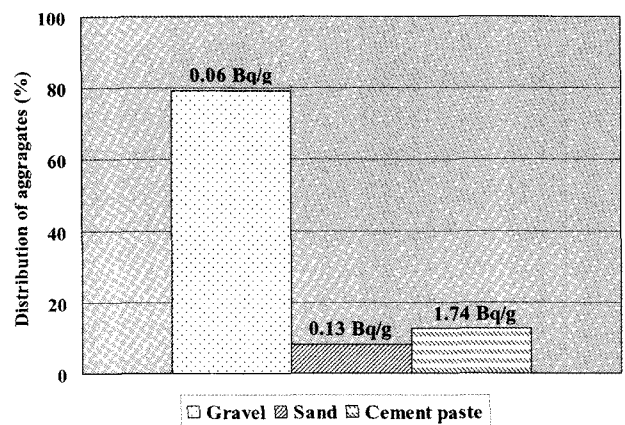


Fig. 7. Distribution of the Aggregates in Activated Heavy Weight Concrete Wastes by Thermal and Mechanical Treatment

mechanical treatment. The initial specific activity of the gravel aggregate measured was 0.17 Bq/g. With the thermal and mechanical processes, the specific radioactivity of the gravel aggregate was considerably decreased by 0.06 Bq/g. A ⁶⁰Co radionuclide was concentrated in the cement paste. The ⁶⁰Co is an important radionuclide in the D&D field. In particular, the major nuclide in the activated heavy weight concrete generated from KRR-2 was ⁶⁰Co. Based on this experiment, the gravel aggregate of low specific activity was obtained at about 80% of activated heavy weight concrete wastes generated from KRR-2. In 1952, Kalousek proposed that all Fe³⁺ ions present in cement eventually enter C-S-H [29]. Copeland et al. [30] provided some support to this theory by suggesting that two Fe³⁺ ions replace one calcium and one silicon. Tits et al. [31] described the adsorption of Eu (III) on C-

S-H. The results showed that the sorption process controlling the retention of Eu is fast, indicating that uptake is probably controlled by an adsorption process. The sorption mechanisms of Eu on the C-S-H phases of hydrated cement were investigated by Pointeau et al. [32]. The Ca:Si ratio corresponding to aged cements was chosen for the C-S-H phases in order to simulate long-term behavior. It was strongly retained on the C-S-H (more than 99.8% sorbed). The binding capacity of cement for Cs has been found to be dependent on the chemical composition, as the different phases have different binding capacities [33]. Cs retention has been found to increase with an increasing CaO:Al₂O₃ ratio. Noshita et al. [34] found that the Cs was highly sorbed by calcium silicate compounds. This suggests that sorption was mainly determined by Ca:Si ratios. Iwaida et al. [35] observed a shortening of the silicate chains in the C-S-H that sorbed Cs. This suggests that hydroxide played an important role in this process. As noted above, most of the nuclides that combined with C-S-H became concentrated into cement paste. When a thermal decontamination process is used, heat becomes a crucial variable for removing the cement paste from the concrete waste because the heat breaks the C-S-H chain structure, which weakens the adherence force between the aggregate and cement matrix combined with nuclides.

3.3 Uranium Contaminated Light Weight Concrete Waste

Two types of concrete wastes are produced from a uranium conversion plant, such as mortar cement concrete waste and general light weight concrete waste. Uranium can form calcium urinate or calcium-uranium-silicate in concrete, depending in the availability of silica [22]. Particulate contamination by U may be present in the form of actinide-IV oxides, such as UO₂.

Figure 8 shows the distribution of the aggregates in uranium contaminated light weight concrete waste by thermal and mechanical treatment. The general light weight concrete could be separated into gravel and sand aggregates of more than 75% with a low specific radioactivity of gravel aggregate at 0.28 Bq/g. The initial specific activity of the light weight concrete waste was 6.5 Bq/g. The uranium was removed easily from the light weight concrete wastes by heating, which weakens the adherence force between the cement matrix and aggregates by mechanical treatment. It is possible to reduce the volume reduction of light weight concrete waste contaminated with uranium compounds generated from UCP more than 100 tons. The main source of cohesion in cement paste is the nanoparticles of C-S-H, which are formed upon the dissolution of the original tricalcium silicate (C₃S). The interaction between highly charged C-S-H particles in the presence of divalent calcium counter ions is strongly attractive because of ion-ion correlations and a negligible entropic repulsion. The reactions between CaO, SiO₂, and UO₂·2H₂O

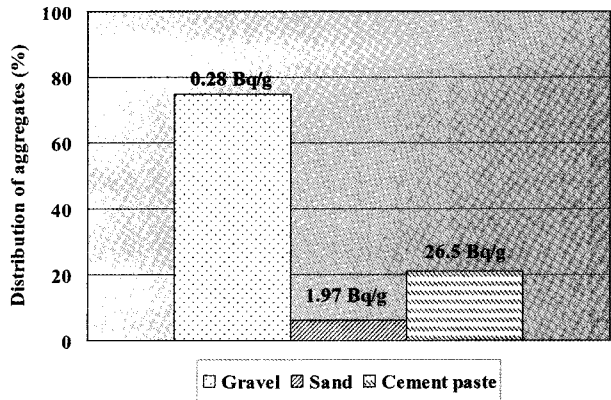


Fig. 8. Distribution of the Aggregates in Uranium Contaminated Light Weight Concrete Wastes by Thermal and Mechanical Treatment

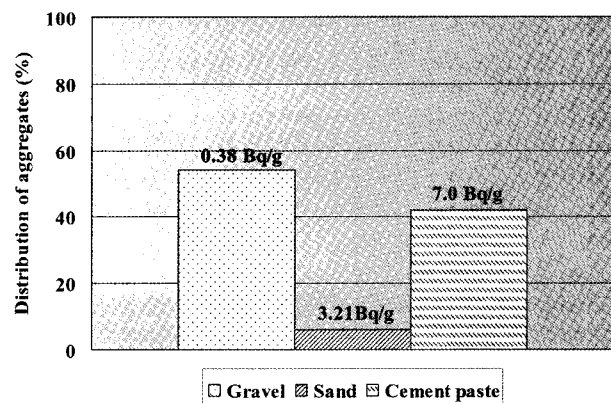


Fig. 9. Distribution of the Aggregates in Contaminated Uranium Mortar Cement Waste by Thermal and Mechanical Treatment

have been studied by Moroni and Glasser [36] in aqueous suspensions at 85°C. Several solubility-limiting, but hitherto uncharacterized, CaO-UO₃-SiO₂-H₂O phases were described. It was concluded that weeksite-like becquerelite-like structures probably developed. Sylwester et al. [37] have shown that U remains in the form of UO₂²⁺. Evidence of inner-sphere interactions on both treated and untreated cements at all pH values was found, with UO₂²⁺ complexing with the mineral surface via sharing of equatorial oxygen. On hydrothermally treated cement, near-neighboring U-U interactions were also observed, indicating the formation of oligomeric surface complexes, or surface precipitates. A study by Pointeau et al. [38] showed that the uptake of U(IV) increased when comparing the least and most degraded cement pastes, whereas the initial composition of the cement had relatively little effect. The coordination environment of U(IV) in the sorption samples was independent of the C-S-H synthesis procedure [39]. The coordination environment of U (IV) in the co-precipitated

samples depended on the synthesis method, and the spectra differed from those for the sorption samples. Min et al. [40] showed that capturing the uranium increased at the high silica phase, regardless of contamination level.

Figure 9 shows the distribution of aggregates in uranium contaminated mortar cement by thermal and mechanical treatment. The initial specific activity of the mortar cement was 4.2 Bq/g. The specific radioactivity of the collected gravel aggregate was decreased by 0.38 Bq/g. Mortar cement of about 54% could be treated with a low specific radioactivity. The recovery rate of the gravel aggregate was lower than that of the light weight concrete waste. The cement paste of the mortar cement was contaminated by the uranium inorganic compounds (AUC, ADU and UO_2) of the particulate.

4. CONCLUSIONS

The characteristics of the separation of aggregates and the distribution of the radioactivity into the aggregates for volume reduction by using activated heavy weight concrete and uranium contaminated light weight concrete waste by thermal and mechanical processes were investigated. Through these experiments, the following conclusions can be drawn:

- Radionuclides were easily removed from the concrete waste by thermal treatment to weaken the adherence force between the cement matrix and the aggregate, followed by mechanical treatment.
- Radioactivity was mainly concentrated in the porous cement paste.
- The specific activity of the aggregates was influenced by the heating temperature and aggregate size.
- A volume reduction of the activated heavy weight concrete waste and uranium contaminated light weight concrete waste was achieved by up to about 80% and 75%, respectively.

It is expected that the volume of radioactive concrete wastes generated from the KRR-2 and UCP can be reduced effectively by thermal and mechanical treatment.

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