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Study of the Irradiation Effect on Waste Forms: A Review

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1. Introduction

The safe disposal and long-term storage of radioactive waste is a major challenge with significant implications for environmental protection and human health. Radiation emitted from radioactive waste can affect the structural stability of solidified materials. Therefore, durability of the solidified material is essential.

In the case of Republic of Korea (ROK), waste form containing organic radioactive waste, such as ion exchange resins (IER), should meet the waste acceptance criteria (WAC) for long-term safety. In this situation, gamma irradiation tests should be conducted at a minimum dose of 10^6 Gy. This dose corresponds to the radiation exposure from Cs^{137} or Sr^{90} at a concentration of 10 Ci/ft^3 over 300 years. Due to the complex chemical composition of these wastes containing IER and the potential for structural changes upon irradiation by radionuclides, the gamma irradiation test of the WAC is essential. However, some countries (France, Japan, etc.) indicate different WAC for the irradiation test.

Therefore, this study reviews the results and mechanisms of existing studies on the irradiation effect for various waste forms (OPC and geopolymer). As a result, this review can be contributed to optimizing the ROK standard for gamma irradiation test.

2. Solidification Materials

2.1. Ordinary Portland Cement [1]

Ordinary portland cement (OPC) is one of the most commonly used materials for the solidification of radioactive waste. In general, tricalcium silicate ($3CaO \cdot SiO_2$) is the main component in the clinker of OPC. It reacts with water to produce calcium silicate hydrate (C-S-H) through a hydration reaction.

However, OPC is produced by heat treatment of limestone ($CaCO_3$) at about 1,400°C, causin g CO_2 and air pollution. As a result, various research has been conducted to explore alternative materials with lower environmental impact.

2.2. Geopolymer [2]

Geopolymer is an inorganic alkali activated material, based on polycondensation reaction in which an aluminosilicate raw material reacts with a strong alkali solution to form a three-dimensional amorphous gel. The formation process of the geopolymer is shown in Fig. 1. For the formulation of geopolymers, raw materials containing high ratio of aluminosilicates, such as metakaolin, are required. In addition, NaOH or KOH are commonly used as alkaline activators.

Compared to OPC, geopolymer can offer several advantages. It is environmentally friendly with significantly reduced CO_2 emissions during formulation. In addition, geopolymers are highly durable and can maintain excellent stability under acid and/or high temperature conditions.

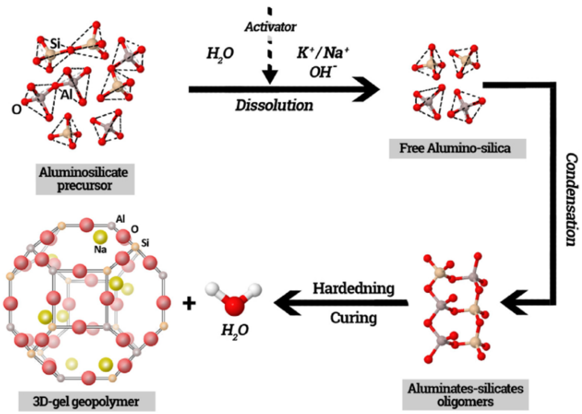

Fig. 1. General geopolymerization mechanism [2].

3. Irradiation test of the WAC

WAC is an essential condition for safety disposal. In this WAC, the irradiation test should be conducted to evaluate the structural changes of waste forms containing organic radioactive waste.

As shown in Table. 1, the current irradiation test standard of ROK is based on documents from the U.S. In contrast, other countries (France and Japan) indicate the different standard. Therefore, we need to optimize the irradiation test standard through basic information from the various recent research.

Table. 1. Irradiation test conditions and assessment criteria depending on the country [3].

Country	Irradiation dose	Assessment criteria
ROK	10^6 Gy (Case: organic waste)	Compressive strength ($\geq$3.445 MPa) measurement
U.S.	10^7 Gy (Case: inorganic waste)	
Japan	10^5 Gy	Damage confirmation
France	2×10^2 Gy/h (Total 10^5 Gy)	Compressive strength change ratio < 20%

4. Effect of radiation

4.1. Effect of radiation on the OPC [4]

In the previous study by Bykov., et al., the structural changes and mechanical strength after gamma irradiation on OPC were analyzed. The compressive strength within the irradiation dose range of 0.4 to 3.2 MGy showed no significant changes. XRD and XPS analyses were conducted to investigate the effects of gamma irradiation on the main chemical composition of OPC; specifically, an increase in the concentration of $CaCO_3$ and a decrease in the C_2S phase were analyzed. SEM-EDX analysis showed minimal microstructural change in the OPC after irradiation.

Furthermore, as shown in Fig. 2, analysis of the pore structure revealed a decrease in porosity due to a decrease in pore size, which may positively impact on long-term mechanical stability. The results of this study demonstrate the potential for OPC to serve as a durable matrix for the solidification of radioactive waste.

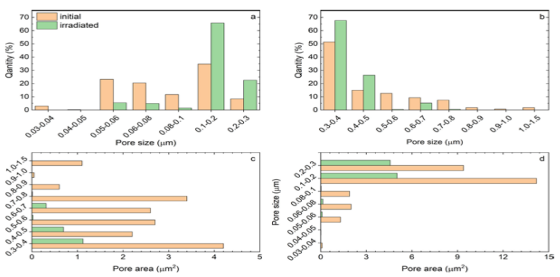

Fig. 2. Pore size (a, b) and pore area (c, d) distributions of OPC with and without irradiation for pore sizes of 0-0.3 μm and ≥0.3 μm, respectively [4].

4.2. Effect of radiation on the geopolymer [5]

In the previous study by Lambertin, D., et al., the effect of gamma irradiation on the structure of Na-based geopolymer was analyzed. As shown in Fig. 3, the results showed that the compressive strength increased by about 10% after gamma irradiation at 50, 500 and 1,000 kGy.

Nitrogen adsorption analysis indicated that the overall volume of pores remained nearly unchanged, but the pore size distribution changed due to the formation of new pores of 7 nm after irradiation. X-ray pair distribution function analysis confirmed that the network structure was changed to denser. These results suggest that gamma irradiation improves the strength and structural stability of geopolymer matrix.

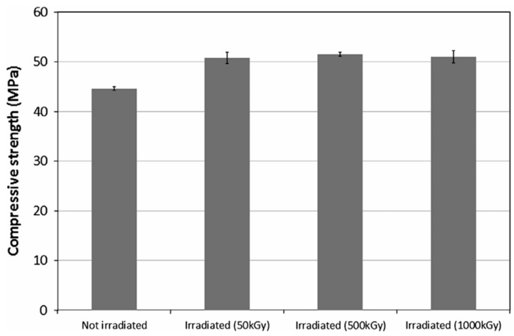

Fig. 3. Compressive strength of geopolymer samples with gamma irradiation exposure at 0, 50, 500 and 1,000 kGy [5].

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