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A Study on the Long-Term Integrity of Polymer Concrete for High Integrity Containers

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During the operation of a nuclear power plant (NPP), the generation of radioactive waste, including dry active waste (DAW), concentrates, spent resin, and filters, mandates the implementation of appropriate disposal methods to adhere to Korea's waste acceptance criteria (WAC). In this context, this study investigates the potential use of polymer concrete (PC) as a high-integrity container (HIC) material for solidifying and packaging these waste materials. PC is a versatile composite material comprising binding polymers, aggregates, and additives, known for its exceptional strength and chemical stability. A comprehensive analysis of PC's long-term integrity was conducted in this study. First, its compressive strength, which is crucial for ensuring the structural stability of HICs over extended periods, was evaluated. Subsequently, the resilience of PC was tested under various stress conditions, including biological, radiological, thermal, and chemical stressors. The findings of this study indicate that PC exhibits remarkable long-term properties, demonstrating exceptional stability even when subjected to diverse stressors. The results therefore underscore the potential viability of PC as a reliable material for constructing high-integrity containers, thus contributing to the safe and sustainable management of radioactive waste in NPPs.

Keywords: Polymer concrete, High integrity container, Long term stability

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1. Introduction

During the operation of a nuclear power plants (NPP) various forms of radioactive waste are generated [1]. These are categorized into dry active waste (DAW), spent resin, boron concentrates, spent filters, etc. The spent resin and concentrates are generated during the water-related treatment processing [2]. One widely used purification method involves ion exchange resins (IER). Since IERs capture radioactive nuclides and impurities in the coolant, they are usually classified as intermediate-level waste (ILW) and/or low-level waste (LLW). After adequate purification, the IER is periodically replaced with new IER to maintain the purification quality [3]. The removed spent IER is sent to the spent resin storage tank and stored temporally. IERs from the chemical and volume control system (CVCS) and spent fuel pool cooling and cleanup system (SFPCCS) are usually classified as ILW or LLW. IERs from the liquid radioactive waste system (LRS) and steam generator blowdown demineralizer (SGBD) are classified as LLW or very low-level waste (VLLW).

Spent IER usually requires conditioning and/or treatment to satisfy the waste acceptance criteria (WAC) [4]. The most promising treatment technology for spent IER is high-integrity container (HIC) packaging, which involves serial treatment of dewatering, drying and packaging in the HIC. The combination of a spent resin drying system (SRDS) and HIC is widely adopted in current NPPs in Korea. The WAC for caverns indicates that spent IER and concentrate shall be solidified or confined in HICs. The solidified radioactive waste should satisfy the requirements, which include sufficient compressive strength, radiological stability, thermal cycle stability, etc. The packaging of concentrate and spent IER allows for simple processing and reduces worker radiation exposure compared to the solidification process. The solidification process usually involves manual transportation and operation of the equipment, so the workers are exposed to external radiation during the processes.

Various HICs have been studied and applied internationally due to its flexibility in processing to satisfy the WAC. A HIC is defined as a container that maintains its integrity for more than 300 years under generic underground environment and disposal conditions [5, 6]. The HIC offers flexibility in required properties, such as free-standing water ratio requirement in radioactive waste, exemption of solidification for concentrates and spent resin, etc.

Polymer concrete (PC) is a multi-composite material consisting of a binding polymeric material, aggregates, and additives [7]. Compared to the conventional, ordinary concrete, the cement hydrate binder is replaced with polymer binding materials. PC offers various advantages: a rapid fabrication process, good water/gas permeation resistance, good adhesion, high chemical resistance, etc [8]. Due to its exceptional advantages, PC is widely applied in various industrial applications.

It has also been noted that PC exhibits exceptional characteristics when considering radiation exposure scenarios. External radiation cause radiation polymerization of monomers and results in a higher level of radiation crosslinking. This indicates that PC has sufficient potential for the application to radioactive waste containers for packaging and disposal [9].

For this paper, the long-term integrity of PC was systematically studied. The mechanical stability, biological stability, thermal stability, chemical stability, and radiological stability were evaluated to understand the characteristics of PC.

2. Methods

2.1 Mechanical Long-Term Stability: Creep Test

The mechanical long-term stability of fabricated PC was studied using a creep test [10]. The diameter and height of the samples, which were fabricated in accordance with

Table 1. Basic characteristics of polymer concrete

Compressive strength (MPa)	Tensile strength (MPa)	Density ($kg \cdot m^{-3}$)	Elastic modulus (MPa)	Poisson ratio
120	10	2,300	32,161	0.18

KS F 2403:2019, were 150 mm and 300 mm. The samples were loaded into the creep test facility. The strain of the polymer concrete was characterized using was strain gauges. The strain was measured for 365 days with loading of 48 MPa, which is 40% of the compressive strength of pristine polymer concrete.

2.2 Biological Stability

2.2.1 General Microorganism Stability

The general microorganism stability was studied using ASTM G21. Aspergillus brasiliensis ATCC 9642, Aspergillus pullulans ATCC 15233, Chaetomium globosum ATCC 6205, Penicillium funiculosum ATCC 11797, and Trichoderma virens ATCC 9645 were used. After cultivation of the microorganisms in the batch with polymer concrete for four weeks at 35℃, the properties of the polymer concrete were analyzed.

2.2.2 Sulphate Microorganism Stability

The sulphate microorganism stability was studied using KCTC 2845 Starkeya novella (S. novella) and KCTC 2846 Paracoccus versutus (P. versutus). After cultivation of the microorganisms in the batch with polymer concrete for 12 weeks at 35℃, the properties of the polymer concrete are analyzed.

2.3 Radiological Stability

The monolithic PC was exposed to 1.0×10^7 Gy of gamma rays using a Cobalt-60 irradiator (MDS Nordion INC, Canada) at the Korea Atomic Energy Research Institute (KAERI, Korea). After irradiation of the PC with gamma rays, the compressive strength test was measured.

2.4 Freeze-thaw (Thermal Cycle) Stability

The physical properties of the polymer concrete were evaluated within the range of −18 to +4℃ (300 cycles) in accordance with the standard method in KS F 2456:2018.

2.5 Chemical Stability

The chemical solutions 10% citric acid, 10% EDTA, 2% HCl, 2% H_2SO_4, and 2% NaOH, were prepared to evaluate the long-term chemical stability of polymer concrete. The polymer concrete specimens were immersed in the chemicals for 1.5 months, 3 months, 6 months, and 9 months at 20 ± 5℃. After immersion in the chemicals, the polymer concrete specimens were washed with distilled water and dried naturally. Their compressive strength was then measured.

3. Results and Discussions

3.1 Basic Properties of Polymer Concrete

Polymer concrete, which is composed of polymer, coarse/fine aggregates, and additives, exhibits good mechanical properties, as shown in Table 1. The microstructure of PC is shown in Fig. 1. It was found that the aggregates and additives, which are primary substances of PC, combined well with the adhesive polymer matrix without significant vacancy or fracture. The Fig. 2 indicates that the polymeric material combined with aggregates tightly,

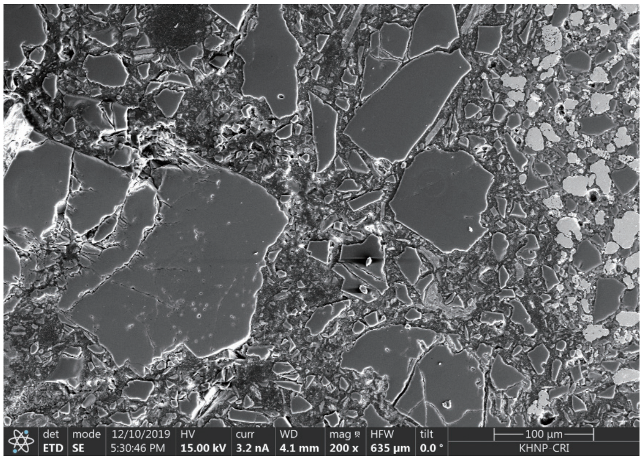

Fig. 1. SEM image of pristine polymer concrete.

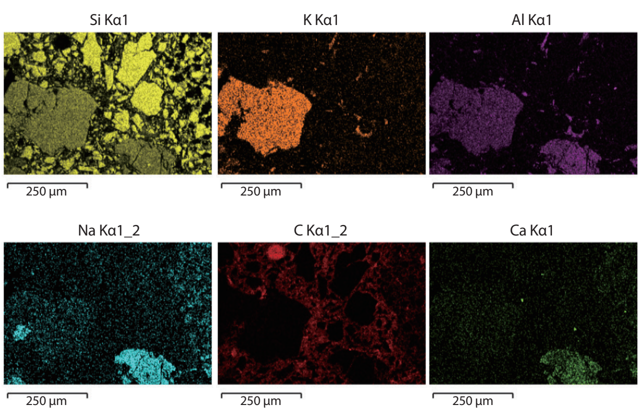

Fig. 2. Element x-ray mapping of the pristine polymer concrete.

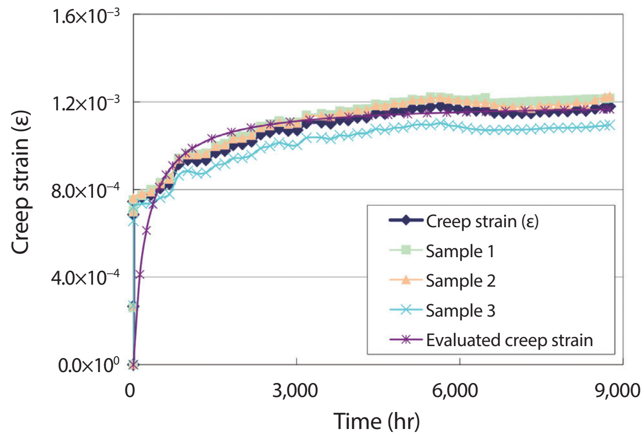

Fig. 3. Measured and evaluated creep stain of the polymer concrete.

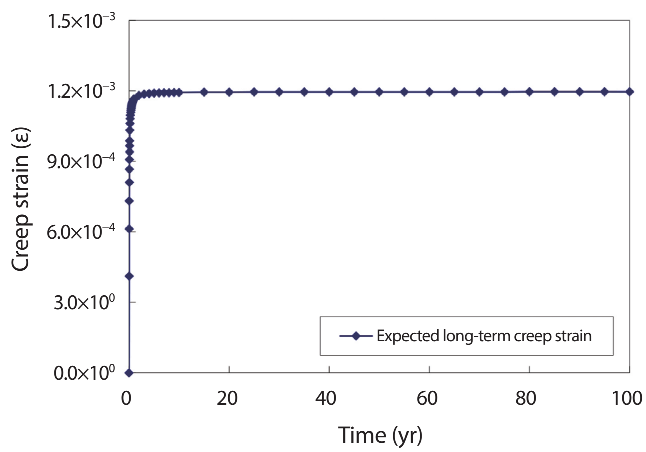

Fig. 4. Expected long term creep strain using Ross equation.

without any vacancy and bubbles. The clear distribution of carbon related chemical species and aggregates, composed of Si, K, and Al, is observed. The Poisson ratio, ratio of the change in the width per unit width of a material, to the change in its length per unit length, as a result of strain, is shown in Table 1 [11]. The Poisson ratio of PC is similar to the ordinary concrete [12].

3.2 Mechanical Long-Term Stability: Creep Test

Fig. 3 shows the one-year creep test results for polymer concrete with loading of 48 MPa and 40% pristine polymer concrete. The minimum, maximum, and average creep strain were 7.620×10^{-4}, 1.1788×10^{-3}, and 9.7040×10^{-4}, respectively. Fifty percent of the total creep strain takes place within 63 days. It was found that the creep strain increases a lot in the early stage of the test. It was also found that the creep strain becomes saturated as the creep test time increases. The calculated creep coefficient was 0.559.

The Ross equation enables the prediction of creep strain. The derived Ross equation, using experiment data in Fig. 4, is shown below.

$$C = \frac{T}{191{,}043 + 836T} \tag{1}$$

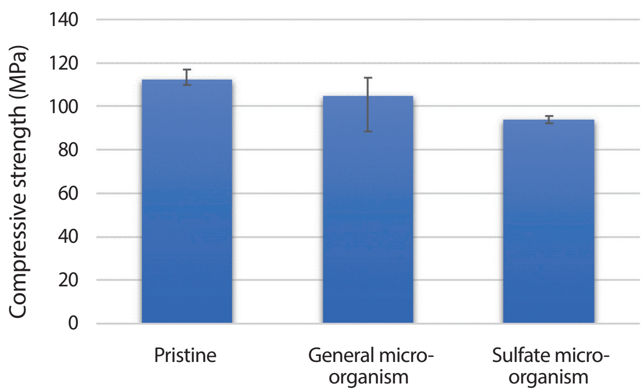

Fig. 5. Compressive strength of pristine and biologically stressed polymer concrete.

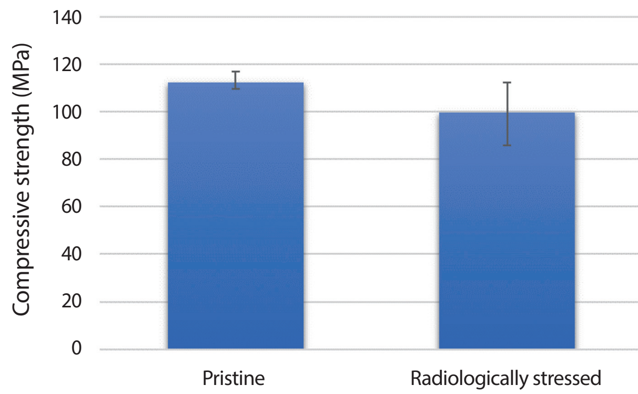

Fig. 6. Compressive strength of pristine and radiologically stressed polymer concrete.

where, C is creep strain and T is time.

The derived Ross equation indicates that the creep strain for 300 years, 600 years, and 1,000 years is 1.1963×10^{-3}, 1.1963×10^{-3}, and 1.1963×10^{-3}, respectively. It was found that the change in creep strain was negligible after a few decades. According to the creep test, it is reasonable to conclude that polymer concrete exhibits good long-term mechanical stability.

3.3 Biological Stability

The general microorganism stability test indicated that the growth of microorganisms at the surface of the polymer concrete was less than 10%. The compressive strength after the general microorganism stability test at four weeks was 104.7 MPa (~15,185 psi), which is similar to pristine polymer concrete. It is reasonable to conclude that polymer concrete is stable under general microorganism growth.

The sulfate microorganism stability test indicated that polymer concrete is stable under microorganism growth. Corrosion of the surface is not observed under 12 weeks of the stability test. The compressive strength after the stability test under S. novella and P. versutus was 94.1 MPa (~13,648 psi). The results indicate that polymer concrete is stable under sulfate microorganism growth. The compressive strength of pristine and biologically stressed PC is shown in Fig. 5.

3.4 Radiological Stability

The material reliability with respect to radiation exposure was evaluated. The compressive strength of polymer concrete is shown in Fig. 6. It was found that the mechanical properties were similar after gamma radiation exposure of 1.0×10^{7} Gy. The resulting compressive strength was 99.8 MPa (~14,475 psi), which is far higher than the requirement of the WAC in Korea. The result indicates that PC is stable under an atmosphere with radiation exposure. It is reasonable to conclude that the mechanical stability is maintained under disposal conditions.

3.5 Thermal Stability

The measured compressive strength of pristine and thermal stressed polymer concrete, shown in Fig. 7, indicates that changes in the properties of the material under thermal stress are negligible. In accordance with the standard test procedure of KS F 2456: 2018, the thermal resistance in the range of −18 to + 4°C was studied for 300 cycles to evaluate changes in the physical properties of polymer concrete. The stress condition appears conservative when

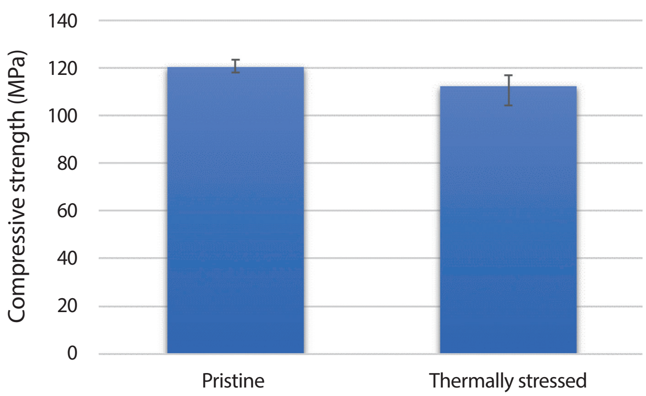

Fig. 7. Compressive strength of pristine and thermally stressed polymer concrete.

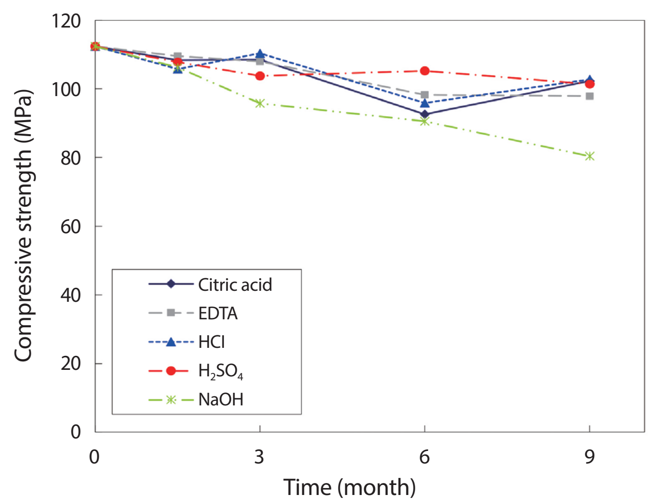

Fig. 8. Compressive strength of chemically stressed polymer concrete.

considering the actual maximum average temperature of the caverns used for radioactive disposal sites is 19°C. It is generally accepted that the temperature of the silo, located in 130 m under the ground, is stable and maintains almost room temperature.

3.6 Chemical Stability

The compressive strength of chemically stressed polymer concrete is shown in Fig. 8. It was found that polymers that were chemically stressed under citric acid, EDTA, HCl and H_2SO_4, exhibit similar mechanical properties until nine months. The compressive strength of polymer concrete stressed under basic conditions and placed in NaOH solution gradually decreased with respect to the stress time. After nine months of stress under an NaOH atmosphere, the compressive strength decreased to 80 MPa (~11,603 psi). It is generally understood that the alkali silica reaction (ASR) takes place in cement concrete under high pH conditions (pH > 13.5). In a cement concrete matrix, the alkali cations react with hydroxyl ions in the cement paste and reactive SiO_2 in the aggregates [13, 14]. Since cement paste is not used to bind the aggregates in PC, the reaction between cement paste and alkali cations does not take place. In a PC matrix, a polymer binder combines the aggregates and additives. It seems that unreacted hydroxyl groups in the aggregates and additives participates in the ASR in PC. The results of the chemical stability tests indicate that PC is far more stable compared to conventional cement concrete.

4. Conclusion

Polymer concrete is fabricated with aggregates, additives, polymers, etc. The basic and long-term mechanical properties were studied. The results indicate that the polymer concrete exhibits good mechanical properties after a long period of use. Various stress condition, including biological stress, radiological stress, thermal stress, and chemical stress, were considered to understand the long-term stability of the materials. The systematic stress tests show that PC exhibits good long-term properties and offers exceptional stability under various stress conditions.

Conflict of Interest

No potential conflict of interest relevant to this article was reported.

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