

Article

Deformability of Bisphenol A-Type Epoxy Resin-Based Polymer Concrete with Different Hardeners and Fillers

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Abstract: This study experimentally investigated the deformability characteristics of bisphenol A-type epoxy resin-based polymer concrete produced using two types of hardener and four types of filler. In particular, the basic properties of epoxy resin polymer concrete, including the modulus of elasticity, setting shrinkage, and thermal expansion, were experimentally investigated to obtain basic data for evaluating compatibility and dimensional stability. The properties of the epoxy resin polymer concrete were determined when different types of hardener and filler were employed. Differences in deformability can be identified based on these properties. In the present study, the setting shrinkage, coefficient of thermal expansion, and modulus of elasticity were lowest when fly ash was employed as one of the four fillers. Hence, it is advantageous to use fly ash as a repair material for ordinary Portland cement concrete structures. Therefore, the results of this study will be helpful when selecting the types of hardener and filler needed to tailor the epoxy resin polymer concrete produced to be suitable for a particular application.

Keywords: epoxy resin concrete; deformability; compatibility; dimensional stability; modulus of elasticity; setting shrinkage; thermal expansion

1. Introduction

Concrete is widely used around the world as a basic construction material. However, in order to keep pace with rapid developments in the construction field, there is now a strong demand to modify the properties of concrete to produce new types [1]. Among those recently developed, the most representative construction material is polymer concrete (PC). PC is a composite material that uses a polymer binder with fine or coarse aggregates and fillers. Neither ordinary Portland cement nor water is employed in its production [2].

The curing of PC depends on the chemical reaction of the resin. Usually, exothermic reactions occur, and long polymer chains are generated during the curing process. The flexibility and strength levels are improved by these long polymer chains. Also, the properties of PC depend highly on the types and contents of the resin, shape and size of the aggregate, filler type, and curing conditions [3–5].

Compared to ordinary Portland cement concrete, PC has superior strength, adhesion, waterproofness, chemical resistance, freeze–thaw durability, and abrasion resistance. These desirable properties originate from the polymer replacing Portland cement [6]. Thus, the weakness of Portland cement concrete (its low strength, poor chemical resistance, and easy cracking) can be prevented by the use of PC [7].

PC is widely employed as a repair material in infrastructure such as concrete structures, pavement, etc. In addition, it is often used in concrete utility systems such as acid waste outlets, underground storage, junction boxes, sewer pipes, and so forth. Plus, it is frequently used for transportation-related concrete structures such as median barriers, bridge panels, railroad ties, and building panels [6,8–10].

Epoxy (EP) resins, unsaturated polyester resins, vinyl ester resins, and methyl methacrylate monomers are all employed as polymeric binders when PC is produced. Among them, composites using EP have been employed widely for the last several decades as repair materials [2,11,12]. Also, EP is one of the most widely used thermosetting polymers due to its remarkably advantageous chemical and mechanical properties [13]. In particular, EP has been used extensively as an adhesive for attaching FRP strips to masonry or concrete supports [14,15]. However, EP is also employed to produce PC or polymer mortar because of its excellent interfacial adhesion and durability. Hence, PC is popular for use in the repair and rehabilitation of Portland cement concrete structures [16].

Among the EPs that currently exist, bisphenol A is the most commonly used because it has a superior response with epichlorohydrin and is relatively inexpensive compared to other epoxies [17]. However, the cost is still high, and compatibility issues occur because the properties of the epoxy binder and existing concrete substrate are different. Thus, the consumption rate of PC has not increased as was once expected [2,18].

Important properties related to compatibility and dimensional stability include mechanical elements, thermal suitability associated with the coefficient of thermal expansion, modulus of elasticity, creep resistance, rate of strength development, and various curing conditions [19]. Factors affecting compatibility include the type and contents of the EP (i.e., binder), type of filler, use of reinforcing materials such as fiber, and cast and curing temperatures.

Therefore, to consider various conditions associated with the properties of the epoxy resin polymer concrete examined in this study, the present research experimentally investigated the deformability of epoxy resin polymer concrete prepared using bisphenol A epoxy, two types of hardeners, and four different types of fillers.

2. Research Significance

EP concrete (or EP mortar) is widely employed in the repair of existing Portland cement concrete structures, more so than in the construction of precast products. This is due to its physical, mechanical, and chemical properties, as well as cost. The most crucial consideration when selecting a repair material is its compatibility with the material employed in the existing structure [20]. Basic elements associated with compatibility include the modulus of elasticity, setting shrinkage, and thermal expansion. A stress–strain relationship occurs when loads are applied; this relationship is related to the modulus of elasticity. Setting shrinkage begins during the curing process immediately after casting PC; thermal strain occurs repeatedly with any temperature change. These properties are closely related to the compatibility between the parent and repair concretes. If the materials are incompatible, the adhesive layer debonds, degrading the durability of the repair.

In previous studies of PC, researchers simultaneously carried out experiments regarding the modulus of elasticity and setting shrinkage [20], and thermal expansion and modulus of elasticity [21,22]. Other studies [5,18] examined the modulus of elasticity, setting shrinkage, and thermal expansion individually. However, it is difficult to find a study that simultaneously considered all three basic elements.

Therefore, the novelty of this study is that the deformability characteristics of bisphenol A-type EP-based polymer concrete (i.e., the modulus of elasticity, setting shrinkage, and thermal expansion) were simultaneously investigated when different types of hardener and filler were employed. The results of this study can serve as a reference for the basic data necessary to establish compatibility and dimensional stability when using EP polymer concrete as a repair material or in precast products.

3. Experiment Details

3.1. Materials

3.1.1. Epoxy Resin and Hardeners

In this study, bisphenol A-type EP was employed, which was diluted with glycidyl ether; it had a low viscosity. According to the standard test method ASTM C881: Standard Specification for Epoxy Resin-Based Bonding Systems for Concrete [23], the viscosity of this EP was Grade 1. It was of a two-part type, characterized by high reactivity. The properties of the EPs used in this study are shown in Table 1, and the properties of the two types of hardener employed can be found in Table 2.

Table 1. Properties of bisphenol A-type EP.

EEW (g/eq)	Viscosity (cps at 25 °C)	Hy-Cl Content (wt.%)	Specific Gravity (20 °C)	Color (G)
183.1	1020	0.1 max	1.14	0.1

Table 2. Properties of hardeners.

Type	Amine Value (HCl) (mg KOH/g)	Viscosity (cps at 25 °C)	Color (G)
Aliphatic amine modified type (AAMT)	376.8	1200	6.1
Cycloaliphatic amine modified type (CAMT)	366.3	690	0.1

3.1.2. Siliceous Aggregate

A siliceous aggregate was used for this experiment; its properties are shown in Table 3. Also, the grading of the aggregate employed in combination is shown in Table 4.

Table 3. Properties of siliceous aggregate.

Size (mm)	Apparent Density	Bulk Density	Unit Weight (kg/m ³)	Fineness Modulus	Moisture Content (%)
0.2–8.0	2.65	2.61	1652	2.81	<0.1

Table 4. Grading combination for aggregates.

Size (mm)	4–8	2–5	1–2	0.8–1.8	0.4–0.9	0.2–0.4
Percentage of mass	2	12	18	22	25	21

3.1.3. Filler

Four types of filler were used: silica flour (SF), ground calcium carbonate (GCC), and waste materials such as blast furnace slag (BFS) and Type II fly ash (FA). Their properties are shown in Tables 5 and 6, and their particle size distributions are listed in Figures 1 and 2. The filler particle size distributions were determined using a laser scattering particle size analyzer (LS 13 320, Beckman Coulter Inc., Brea, CA, USA). Chemical composition was analyzed by X-ray fluorescence (XRF, Rigaku Co., Akishima, Tokyo, Japan).

Table 5. Physical and chemical properties of fillers from virgin materials.

Filler	Density (g/cm ³)	Particle Size (μm)			Chemical Composition (%)					
		Mean	Median	Mode	SiO ₂	Al ₂ O ₃	CaO	MgO	Fe ₂ O ₃	SO ₃
Ground calcium carbonate (GCC)	2.70	18.18	14.57	28.70	0.62	0.48	53.8	1.21	0.15	0.01
Silica flour (SF)	2.65	31.77	22.74	37.97	98.7	0.52	0.08	0.03	0.18	0.02

Table 6. Physical and chemical properties of fillers from waste materials.

Filler	Density (g/cm ³)	Particle Size (μm)			Chemical Composition (%)					
		Mean	Median	Mode	SiO ₂	Al ₂ O ₃	CaO	MgO	Fe ₂ O ₃	SO ₃
Fly ash (FA)	2.20	22.29	10.75	7.77	49.1	22.7	7.15	1.33	8.31	0.99
Blast furnace slag (BFS)	2.91	13.41	10.10	16.40	29.3	13.3	48.5	2.72	0.53	2.94

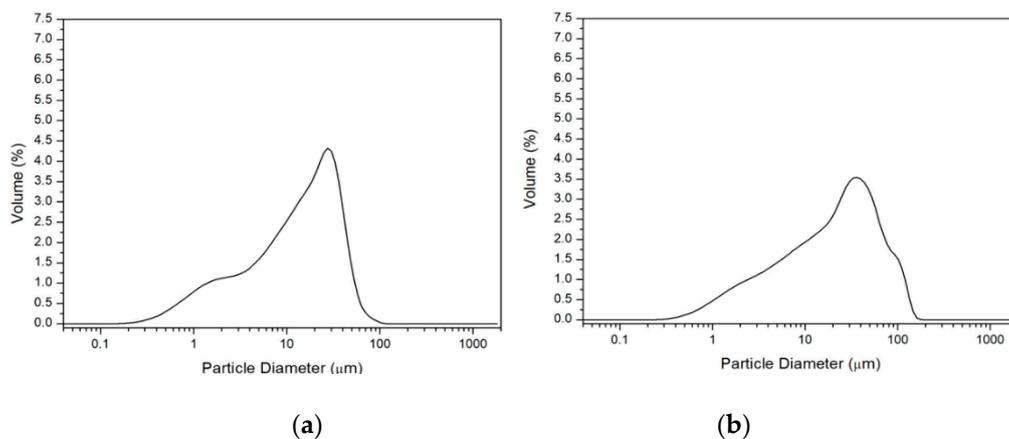


Figure 1. Particle size distribution of fillers from virgin materials: (a) Ground calcium carbonate and (b) silica flour.

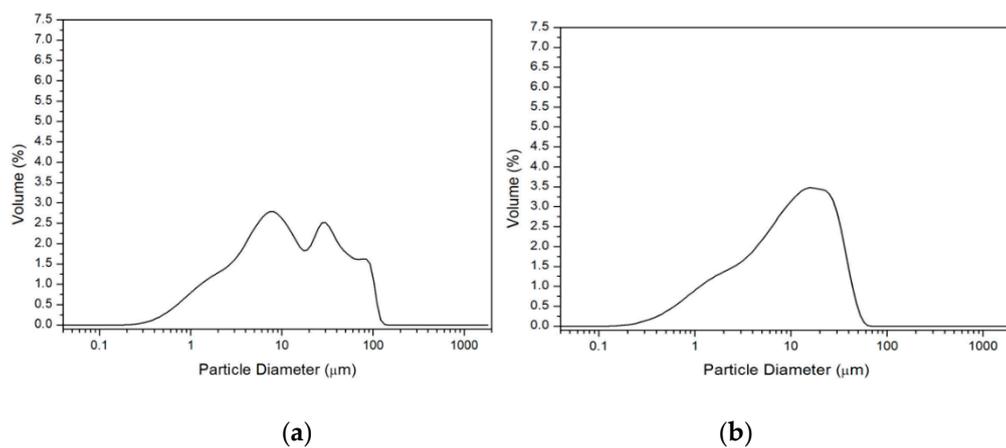


Figure 2. Particle size distribution of fillers from waste materials: (a) Fly ash and (b) blast furnace slag.

3.2. Specimen Preparation

3.2.1. Mix Proportions

Standardized mix design procedures do not, as yet, exist for PC systems produced with binders prepared with aggregates and polymer resins [24]. Hence, in this study, the mix proportions were determined based on a flow of $110 \pm 5\%$, as specified in ASTM C109/C109M-02: testing method for Compressive Strength of Hydraulic Cement Mortar [25]. The optimum polymer content satisfying all

of the mechanical properties for the PC was between 12.4% and 15.6% [26]. In this study, 13% was applied. The mix proportions, as determined by trial and error, are shown in Table 7.

Table 7. Mix proportions of ER concrete (unit: wt %).

Hardener Type	Epoxy	Hardener	Filler	Aggregate
AAMT	9.62 (100)	3.38 (35)	19	68
CAMT	8.97 (100)	4.03(45)	19	68

3.2.2. Mix Description

The specimens were produced through the following process.

- (1) EP and hardening agents were weighed and blended.
- (2) First mix step: The aggregate and filler were placed in the mixer to mix them together.
- (3) Second mix step: The EP and hardener was placed in a container and thoroughly mixed using a stirrer.
- (4) The epoxy binder blended in Step 2 was poured into the mixer that contained the mixed aggregate and filler produced in Step 1, in order to thoroughly mix the prepared epoxy binder with the aggregate and filler.
- (5) The mixed epoxy resin polymer concrete was cast into a metal mold and compacted using a rod and table-type vibrator to void any formation inside the specimens.
- (6) The test specimens prepared via this procedure were either used immediately for the test or after a curing process at room temperature.

3.3. Testing Methodology

3.3.1. Modulus of Elasticity

The modulus of elasticity test was carried out in accordance with ASTM C469M-14: Standard Test Method for Static Modulus of Elasticity and Poisson's Ratio of Concrete in Compression [27]. An electrical resistance strain gauge with a length of 30 mm (PFL-30-11-1LJC, Tokyo Measuring Instruments Laboratory Co., Ltd., Shinagawa, Tokyo, Japan) was attached to each cylindrical test specimen ($\varnothing 50 \times 100$ mm); the strain was measured by a data logger (TDS-602, Tokyo Measuring Instruments Laboratory Co., Ltd., Shinagawa, Tokyo, Japan), as shown in Figure 3. A total of 50 tons was used for loading in the universal testing machine (HD 201, Hyundai Precision Co., Ltd., Daegu, Korea), and a constant rate of 0.25 MPa/s was applied.



Figure 3. Test setup for determining the modulus of elasticity.

3.3.2. Setting Shrinkage

The dry shrinkage test was carried out following ASTM C596-01: Standard Test Method for Drying Shrinkage of Mortar Containing Hydraulic Cement [28]. The test was conducted in a constant temperature/humidity room with a temperature of 23 ± 2 °C and relative humidity of $60 \pm 5\%$. A data

logger (TDS-602, Tokyo Measuring Instruments Laboratory Co., Ltd., Shinagawa, Tokyo, Japan) and embedded-type strain gauge were used to measure the drying shrinkage strain, as shown in Figure 4.



Figure 4. Test setup for measuring drying shrinkage.

3.3.3. Thermal Expansion

The thermal expansion test was conducted according to ASTM E831-14: Standard Test Method for Linear Thermal Expansion of Solid Materials by Thermomechanical Analysis [29]. To measure the thermal strain, a data logger (TDS-602, Tokyo Measuring Instruments Laboratory Co., Ltd., Shinagawa, Tokyo, Japan) and embedded-type strain gauge (PMFL-60-5LT, Tokyo Measuring Instruments Laboratory Co., Ltd., Shinagawa, Tokyo, Japan) were used, as shown in Figure 5. After stabilizing at 25 °C for 5 h, the temperature of the heating oven was increased to 60 °C (5 °C/min) to measure thermal strain.



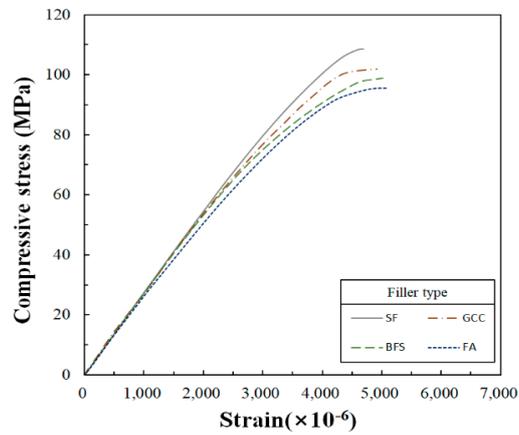
Figure 5. Test setup for measuring thermal expansion.

4. Results and Discussion

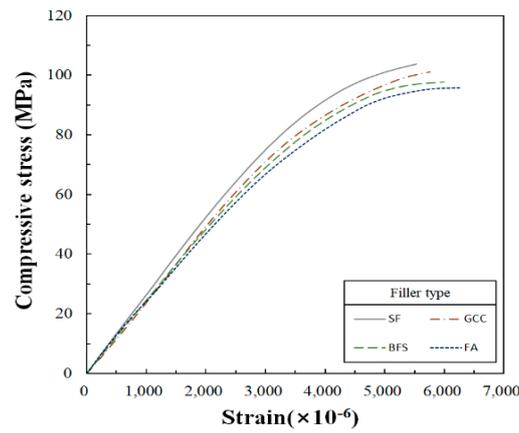
4.1. Modulus of Elasticity

The modulus of elasticity is a material constant determined based on the relationship between stress and strain. According to Hook's law, linear elastic material maintains a constant relationship between stress and strain. Strictly speaking, the elastic modulus section means only the linear section of the stress–strain curve. Hence, the secant modulus of elasticity is applied when the linear section of the stress–strain curve is not straight, such as in the stress–strain relationship of concrete. There are other ways to determine the modulus of elasticity, such as the chord modulus of elasticity and tangent modulus of elasticity, but these do not apply to concrete [30]. The modulus of elasticity is one of the major criteria used to evaluate the deformation characteristics of a structure when a load is applied to structural members. If damaged Portland cement concrete is repaired with an EP polymer concrete mixture, a mismatch in the modulus of elasticity for the two materials will reduce the load-bearing capacity of the combined system [20]. In addition, matching the elastic modulus of the material used for the existing substrate and elastic modulus of the repair mixture is very important in terms of shrinkage and permeability [31]. The stress–strain relationships determined by the compression test in the present study are shown in Figure 6. In this figure, the secant modulus of elasticity was determined from the slope formed by the coordinate origin and point corresponding to 40% of the ultimate stress; the result is shown in Figure 7. The modulus of elasticity ranged from 25.2×10^3 to 27.2×10^3 MPa with the AAMT hardener and from 24.7×10^3 to 27.1×10^3 MPa with the CAMT hardener. From these

results, it was determined that the moduli of elasticity for the two types of curing agent were similar. By filler type, the order from highest to lowest elastic modulus was SF, GCC, BFS, and FA. This order was the same for both types of hardeners. There was a minor difference between the elastic moduli of the two hardener types. The cause for this minor difference is likely related to the mode of particle size employed in this study. To determine the exact cause of this difference, additional research is required.



(a)



(b)

Figure 6. Compressive stress and strain curves by hardener and filler type: (a) AAMT hardener and (b) CAMT hardener.

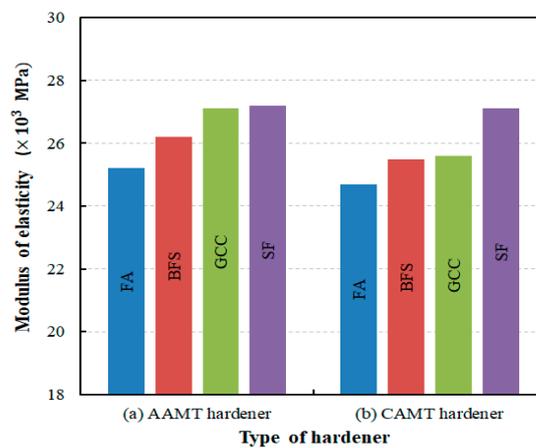


Figure 7. Elastic moduli by hardener and filler type.

According to previous studies on the moduli of elasticity of EP polymer concrete mixtures, Lokuge et al. [5] reported levels ranging from 9.5 to 11 GPa, Hassan et al. [20] showed 13.2 GPa, Decter et al. [31] demonstrated 0.5 to 20 GPa, and Yemam et al. [32] found 8 to 18 GPa. These moduli of elasticity were affected by the type of EP, its contents, the properties of the aggregate, test method employed, temperature, and specimen size. Also, it was determined that the elastic modulus of EP concrete was significantly smaller than the elastic modulus of Portland cement concrete (i.e., 20 to 40 GPa) [6,31]. This low elastic modulus is not disadvantageous in terms of compatibility with Portland cement concrete when the modulus of elasticity of Portland cement concrete is considered [20]. The compressive strengths are shown in Table 8, as determined when failure stresses in the stress–strain curves shown in Figure 6 were regarded as compressive strengths. These compressive strengths were developed at the age of 7 days. By type of hardener, the compressive strengths of the AAMT hardener ranged from 95.5 to 108.6 MPa, and that of the CAMT hardener ranged from 95.8 to 103.8 MPa. From these results, it was determined that there was no significant difference between the two hardeners. For compressive strength by filler type, the order of strength development from highest to lowest was SF, GCC, BFS, and FA. The advantageous property of PC associated with the compressive strength is rapid hardening; about 80% of the 28-day compressive strength can be achieved in seven days [5]. This is an excellent property when the material is used in precast applications.

Table 8. Compressive strengths by filler and hardener type (unit: MPa).

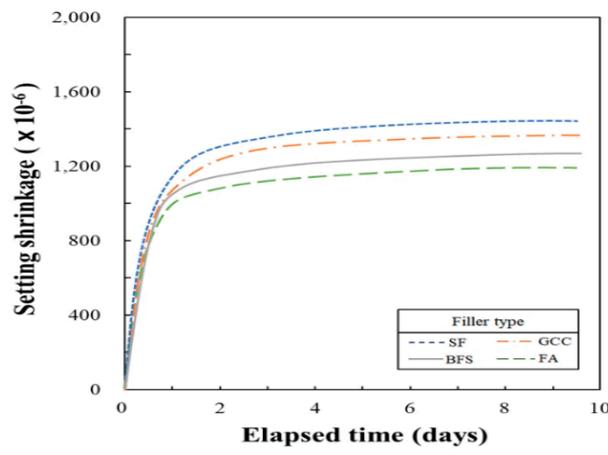
Filler Type	AAMT Hardener	CAMT Hardener
FA	95.5	95.8
BFS	98.9	97.8
GCC	101.9	101.1
SF	108.6	103.8

According to previous studies on the compressive strengths of EP polymer concrete mixtures, Niaki et al. [33] reported a developed compressive strength ranging was from 40.6 to 76.2 MPa, Barbuta et al. [4] found from 43.47 to 69.82 MPa, Hassan et al. [20] showed 88.8 MPa, Bedi et al. [24] determined 65.16 MPa, Decter et al. [31] identified between 50 and 100 MPa, and Yemam et al. [32] indicated 36 to 103 MPa. In particular, Golestaneh et al. [34] showed the highest compressive strength of 128.9 MPa when using silica flour as a filler, which is similar to the test results of this study. The maximum compressive strains obtained from the stress–strain curves seen in Figure 6 are shown in Table 8. By type of hardener, the ultimate compressive strain of the AAMT ranged from 4693×10^{-6} to 5095×10^{-6} and the ultimate compressive strain of the CAMT ranged from 5533×10^{-6} to 6254×10^{-6} . By type of filler, the order of compressive strain from highest to lowest was SF, GCC, BFS, and FA. It was also found that the ultimate compressive strain increased as the elastic modulus and compressive strength decreased.

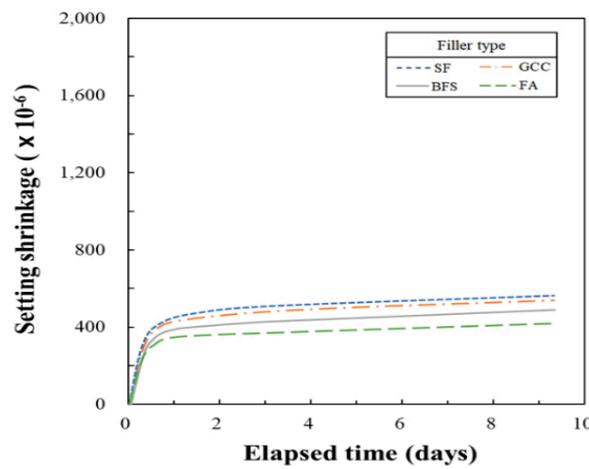
4.2. Setting Shrinkage

Drying shrinkage occurs in ordinary Portland cement concrete mixtures. In contrast, setting shrinkage occurs in PC. This drying shrinkage in Portland cement concrete can be considered similar to the setting shrinkage seen in PC. This setting shrinkage causes shrinkage stress, and cracking occurs when this shrinkage stress exceeds the tensile strength of the Portland cement mixture. Since water is not used in producing PC, its shrinkage mechanism is different. This setting shrinkage is known mainly to be influenced by the type and percentage of polymeric material [7]. Shrinkage is also an important property with regards to the dimensional stability of a combined structure that employs the two different materials. The interface of the combined structure or portions that are low in strength can fail due to internal stresses generated by drying shrinkage. The incompatibility caused by drying shrinkage is greater than that which results from the modulus of elasticity [20]. The test results for setting shrinkage are shown in Figure 8. The setting shrinkage of the AAMT hardener ranged from 1246×10^{-6} to 1497×10^{-6} , and the setting shrinkage for the CAMT hardener ranged

from 403×10^{-6} to 547×10^{-6} , a relatively significant difference (see Figure 9). By filler type, the order from highest to lowest setting shrinkage was SF, GCC, BFS, and FA, and the difference between each setting shrinkage was not significant. The difference by type of hardener was likely due to the various reaction mechanisms according to the hardener’s chemical composition, while the differences between filler type were probably related to the mode of particle size. However, further study is needed to confirm these conclusions. In a previous study, Mani et al. [7] reported that the setting shrinkage of polyester resin concrete ranged from 2900×10^{-6} to 4200×10^{-6} , which was much higher than the setting shrinkage of EP polymer concrete of 1100×10^{-6} . This study also reported that the addition of CaCO_3 micro filler negatively affected the setting shrinkage of polyester concrete. Their results are similar to the test results obtained in the present study. The low setting shrinkage of EP polymer concrete using CAMT hardener is very favorable in terms of compatibility.



(a)



(b)

Figure 8. Variations in setting shrinkage by hardener and filler type: (a) AAMT hardener and (b) CAMT hardener.

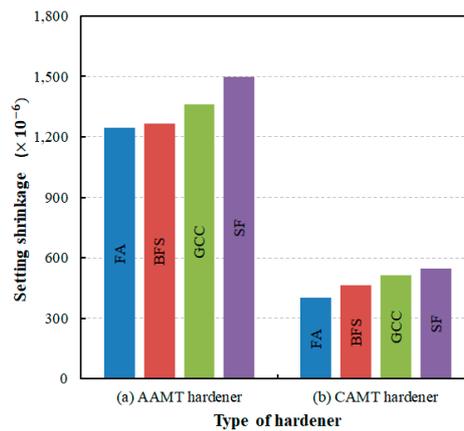
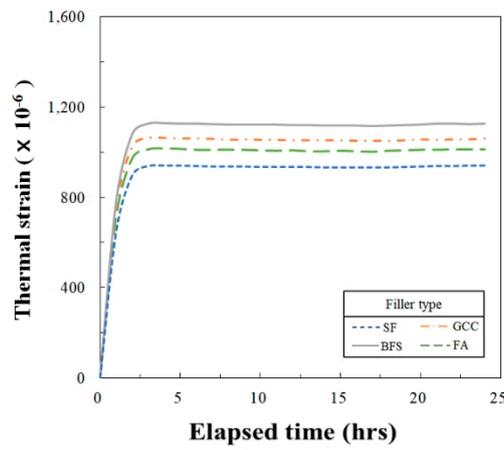


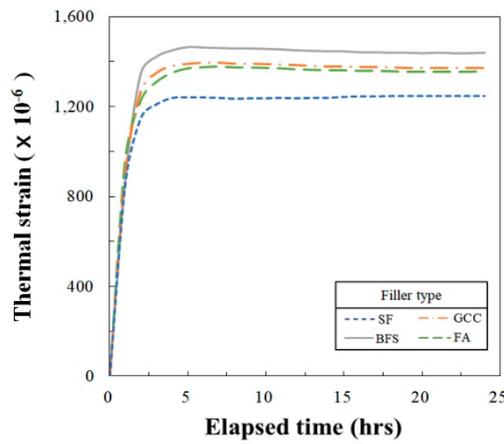
Figure 9. Setting shrinkage by hardener and filler type.

4.3. Thermal Expansion

Thermal expansion and shrinkage occur in ordinary Portland cement concrete. Like ordinary Portland cement concrete, PC also sees thermal expansion, which occurs when the temperature of the hardened mixture rises; thermal shrinkage occurs when the temperature of the hardened mixture decreases. Thermal expansion includes linear and volumetric thermal expansion. The magnitude of the thermal expansion depends on differences in the crystalline and molecular structures of the material [35]. The polymerization of a polymer and changes in temperature are two main elements that change the volume of PC in a structure. If the stress generated by the volume strain exceeds the allowable tensile stress of the PC mixture, cracking will occur. The thermal strain and linear thermal expansion determined in this study are shown in Figures 10 and 11, respectively. By type of hardener, the coefficient of thermal expansion of the AAMT hardener ranged from 18.2×10^{-6} to 22.1×10^{-6} per $^{\circ}\text{C}$, and for the CAMT hardener from 24.3×10^{-6} to 27.4×10^{-6} per $^{\circ}\text{C}$. By filler type, the order from highest to lowest coefficient of linear thermal expansion was SF, GCC, BFS, and FA. The cause of the difference by type of curing agent is likely due to the different chemical compositions. The differences by filler type are likely closely related to the thermal expansion properties of each filler's chemical composition. To determine these causes more accurately, further study is required. According to the results of Decter [31], the coefficient of thermal expansion of EP mortar ranged from 20×10^{-6} to 50×10^{-6} per $^{\circ}\text{C}$, which is larger than the coefficient of thermal expansion obtained in this study. In comparison, the coefficient of thermal expansion of hardened Portland cement concrete is very small. Zoldners [36] reported the value as ranging from 6.3×10^{-6} per $^{\circ}\text{C}$ to 11.7×10^{-6} per $^{\circ}\text{C}$. Decter [31] reported the coefficient of thermal expansion as ranging from 7×10^{-6} to 14×10^{-6} per $^{\circ}\text{C}$. The results of these studies indicate that the coefficient of thermal expansion of epoxy resin polymer concrete is about twice as high as that of ordinary Portland cement concrete [31]; this aligns with the results of the present study. The cause of this significant thermal expansion could be high temperatures resulting from polymerization in the epoxy resin polymer concrete.



(a)



(b)

Figure 10. Variations in thermal strain by hardener and filler type: (a) AAMT hardener and (b) CAMT hardener.

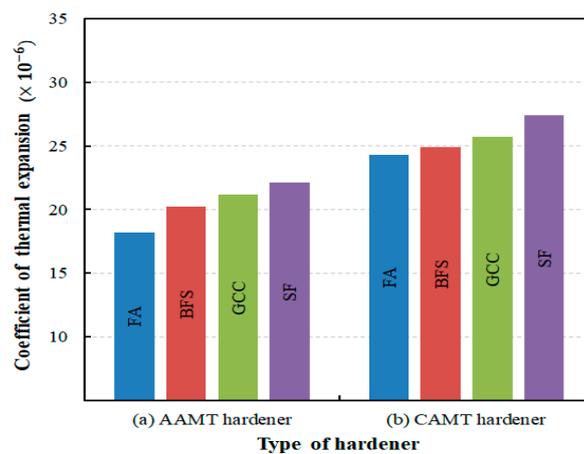


Figure 11. Coefficients of thermal expansion by hardener and filler type.

5. Conclusions

This study experimentally investigated the deformability characteristics of bisphenol A-type epoxy-based polymer concrete by hardener type and filler, including the modulus of elasticity, setting

shrinkage, and thermal expansion. The primary objective of this research was to obtain basic data associated with the compatibility and dimensional stability required to apply EP polymer concrete when repairing damage to concrete structures and producing precast products.

According to these test results, the modulus of elasticity ranged from 25.2×10^3 to 27.2×10^3 MPa when AAMT hardener was employed, and from 24.7×10^3 to 27.1×10^3 MPa when CAMT hardener was used. By filler type, these cases followed the following order: SF, GCC, BFS, and FA. However, there were no significant differences in the modulus of elasticity by type of hardener and filler. The elastic moduli of these EP polymer concretes were significantly lower than the elastic modulus of Portland cement concrete. These low moduli of elasticity are not disadvantageous in terms of compatibility with Portland cement concrete.

The setting shrinkage ranged from 1246×10^{-6} to 1497×10^{-6} for AAMT hardener and from 403×10^{-6} to 547×10^{-6} for CAMT hardener. From this result, it was determined that there was a significant difference in setting shrinkage when different types of hardener were employed. By filler type, the order from highest to lowest setting shrinkage was: SF, GCC, BFS, and FA; the difference between each setting shrinkage was not significant. For the two hardeners employed in this study, it was determined that the low setting shrinkage obtained using the CAMT hardener was more advantageous in terms of compatibility.

The test results by type of hardener indicate that the coefficient of thermal expansion ranged from 18.2×10^{-6} to 22.1×10^{-6} per °C for the AAMT hardener and from 24.3×10^{-6} to 27.4×10^{-6} per °C for the CAMT hardener. In terms of filler type, the order from highest to lowest coefficient of thermal expansion was: SF, GCC, BFS, and FA. In this study, the coefficient of thermal expansion of the EP polymer concrete was twice as high as that of ordinary Portland cement concrete. The cause of this result may be the high temperature generated by polymerization in epoxy resin polymer concrete.

Generally, the deformability of polymer concrete is higher than that of Portland cement concrete. However, considering the four fillers employed in this study, it was determined to be advantageous to use fly ash in the repair of ordinary Portland cement concrete structures because the setting shrinkage, coefficient of thermal expansion, and modulus of elasticity were found to be the lowest. This is a key result of this study.

Basic data for EP polymer concrete, including the modulus of elasticity, setting shrinkage, and thermal expansion, were determined in the present work. Based on this research, further study of the compatibility and dimensional stability of EP polymer concrete is required for field application.

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Conflicts of Interest: The authors declare no conflicts of interest.

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