

Characteristics of Polyester Polymer Concrete using Spherical Aggregates from Industrial By-Products

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ABSTRACT: Despite its excellent physical and mechanical properties, polymer concrete has not been widely used owing to its much higher unit price than conventional portland cement concrete. To ensure the economic efficiency of polymer concrete, it is utmost important to reduce the use of polymer binder, which occupies most of the production cost of polymer concrete. Based on the experimental investigations, replacing filler (calcium carbonate) and fine aggregate (river sand) with fly ash and rapid-cooled steel slag (RCSS), which are spherical materials obtainable from industrial by-products, was found to be effective for improving the strength characteristics and durability as well as the cost efficiency of polymer concrete. The product developed in this study successfully reduced the demand for polymer binder by 21.3% compared to the conventional product, which in turn saved the total material costs by 18.5%. Although the use of RCSS showed performance degradation at an elevated temperature condition, considering typical temperature ranges that actual concrete infrastructures experience, it is expected that the polymer concrete using fly ash and RCSS will provide high-level performances as construction and repair materials. © 2013 Wiley Periodicals, Inc. *J. Appl. Polym. Sci.* 129: 2905–2912, 2013

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INTRODUCTION

Polymer concrete is a composite material incorporating a resin binder instead of portland cement. Research on polymer concrete began in the 1950s and it has been actively used since the 1970s. The excellent performances of polymer concrete composites that are not found in conventional portland cement concrete (such as high strength, water tightness, durability, chemical resistance, wear resistance, and impact resistance^{1–5}) have expanded the usage of polymer concrete for various applications such as floor, pavement, waterproofing, repair, corrosion prevention, and precast products.^{1,6–8} However, to date, the use of polymer concrete has been quite limited owing to its much higher cost than conventional portland cement concrete. Also, the low thermal and fire resistance of polymer concrete is one of the reasons that prevents polymer concrete from being widely used as an alternative material to conventional cement concrete.^{1,9,10} Recently, research has been conducted on the development of polymer concrete using a reduced amount of polymer binder, which accounts for most of the production costs of polymer concrete.¹¹ Among the various types of polymer resin

binders, unsaturated polyester resin is most frequently employed because of its improved performance and economic efficiency.¹²

The primary objective of this study is to examine the effectiveness of replacing calcium carbonate filler by fly ash and river sand by rapid-cooled steel slag (RCSS) in reducing the demand of polymer binders. These spherical materials—fly ash and RCSS—can be obtained as industrial by-products. Fly ash is the ash of coal used in power plants which has a typical particle diameter of $<20\ \mu\text{m}$ and is one of the major industrial by-products that occupies 75–80% of total ash production. On the other hand, RCSS is the spherical aggregate formed by vitrification: rapid cooling of steel slag in fused conditions. This material is also commonly referred to as atomizing steel slag because it is manufactured by spraying high-pressure air.^{13,14} The steel slag is the spherical shape aggregate with various size distributions. Accordingly, it has advantages in enhancing workability by providing a ball-bearing effect when used as alternative fine aggregate for polymer concrete, which can be a potential feature for reducing the demand of polymer binders to obtain the desired workability.

Table I. Physical Properties of Aggregates and RCSS

Type	Source	Density (g/cm ³)	Water content (%)	Unit weight (kg/m ³)	Solid volume (%)	Size (mm)	
Fine aggregate	River sand	Silica	2.62	<0.05	1422	55	0.1–1.2
	RCSS	Steel slag	3.54	<0.05	2263	63.5	0.1–1.2
Coarse aggregate	Silica	2.65	<0.05	1510	52	5–8	

In Korea, about 8 million tons of steel slag is produced every year. The steel slag is different from blast furnace slag as it contains a large amount of free calcium oxide (free CaO) and free magnesium oxide (free MgO). When used in cement concrete, the free CaO and free MgO react with water and then transform to Ca(OH)₂ and Mg(OH)₂, respectively, leading to volume expansion and eventually resulting in cracking.^{15–17} This is the reason why the steel slag cannot be used as fine aggregate for conventional cement concrete. However, as polymer concrete uses a polymer binder instead of water and cement, steel slag can be effectively utilized as fine aggregate for polymer concrete without any potential volume instability problems.

In this study, various specimens with different addition ratios of polymer binder and replacement ratios of RCSS were fabricated, and then compressive and flexural strengths were measured to find the optimum compositions of polymer concrete using fly ash and RCSS. Along with the strength characteristics, total pore volume, porosity, average pore diameter, bulk density, and microstructure were also analyzed using a mercury intrusion method and scanning electron microscope (SEM) because the durability of a material is closely related to the characteristics of pore systems.⁸ Moreover, to examine the possible performance degradation of polymer concrete using fly ash and RCSS under the elevated temperature condition, a hot water resistance test was performed, and then the variations of strength and pore characteristics before and after the hot water resistance test were evaluated.

EXPERIMENTAL

Materials

Unsaturated polyester resin was used as a polymer binder for polymer concrete composite. As the shrinkage reducing agent, polystyrene, a typical crosslinking agent for unsaturated polyester resins, was employed. Methyl ethyl ketone peroxide (MEKPO) and cobalt octoate were used as a catalyst and accelerator, respectively. Fly ash with the diameter <20 μm was used as filler in the place of heavy calcium carbonate. Also, RCSS with the diameter of 0.1–1.2 mm was used as fine aggregate instead of river sand with different replacement ratios. For coarse aggregate, dried crushed stone with the diameter of 5–8 mm was used. Physical properties and chemical compositions of the materials used in this study are summarized in Tables I–III, respectively.

Methods

Compressive and Flexural Strength Tests. Prismatic specimens (60 × 60 × 240 mm) were fabricated in accordance with the KS F 2419 (*Standard test method for making polyester resin concrete specimens for strength test*). Compressive and flexural

strength tests were performed in accordance with the KS F 2481 (*Method of test for compressive strength of polyester resin concrete*) and KS F 2482 (*Method of test for flexural strength of polyester resin concrete*) standards, respectively. For the flexural strength test, a third-point loading method was used.

Absorption Test. The fabricated specimens were cured for 14 days and then they were dried at 80 ± 2°C for 48 h. Subsequently, the weight of the specimens was measured. To determine the absorption rate, the weight of the specimens was measured again after 48 h of absorption test at 20 ± 2°C. The absorption rate was determined as follows:

$$A = \frac{W_1 - W_0}{W_0} \quad (1)$$

where A is the absorption rate (%), W_0 is the weight of a specimen before the test (g), and W_1 is the weight of a specimen after the test (g).

Hot Water Resistance Test. A hot water resistance test was performed by submerging the specimens in 90°C water for 28 days. After the exposure to hot water, the specimens were stabilized and dried at ambient condition for 7 days and then compressive and flexural strengths were measured in accordance with the KS F standards to compare with those measured prior to the hot water resistance test.

Pore Analysis. The samples with the particle size of 2.5–5 mm were collected and cleaned with acetone. After 48 h of drying, pore analyses (i.e., total pore volume, porosity, average pore diameter, and bulk density) were performed using a mercury intrusion method. To analyze the pore characteristics, AutoPore 9505 was employed in this study.

SEM Observation. To observe the microstructure and the bond characteristics between the polymer matrices and the RCSS particles, a high-resolution field emission SEM (Model; MIRA

Table II. Physical Properties of UP Resin and Shrinkage-Reducing Agent

Type	Specific gravity	Nonvolatile materials	Acid value	Viscosity (Poise)
Unsaturated polyester resin	1.1	62	20	4.1
Shrinkage-reducing agent (polystyrene)	0.95 ± 0.002	35 ± 2	-	33

Table III. Chemical Compositions of Fly Ash and RCSS

Materials Component	Chemical compositions (wt %)									
	SiO ₂	Al ₂ O ₃	Fe ₂ O ₃	CaO	MgO	Na ₂ O	K ₂ O	SO ₃	TiO ₂	Others
Fly ash	57.48	24.31	7.39	3.17	1.15	0.57	1.02	2.9	1.18	0.83
RCSS	13.0	7.96	30.2	39.9	8.0	-	0.03	0.1	-	0.66

LMH II) was used. The samples were platinum coated in the vacuum condition after treating with acetone and drying for a sufficient period.

Mixture Proportions

The mixture proportions of polymer concrete used in this study are summarized in Table IV. The polymer binder was prepared by mixing 80 wt % of unsaturated polyester resin and 20 wt % of shrinkage reducing agent. Four different addition ratios of polymer binder were used, that is, 7.5, 8.0, 8.5, and 9.0 wt %. The addition ratio of fly ash replacing calcium carbonate filler was selected to fill the same volume fraction with the calcium carbonate filler (i.e., 20%). The replacement ratios of RCSS replacing fine aggregate (river sand) were 0, 25, 50, 75, and 100 vol % and the volume of RCSS was also set to be the same as

that of the fine aggregate. The addition ratio of coarse aggregate varied between 51 and 52.5% to fabricate total 20 specimens with different mixture proportions. All the specimens were cured at room temperature ($20 \pm 2^\circ\text{C}$) for 14 days.

RESULTS AND DISCUSSION

Absorption Rate

Figure 1 shows the variations of absorption rate depending on the RCSS replacement ratio and the polymer binder addition level. As the result shows, the absorption rate tended to decrease as the addition amount of polymer binder increased. This is most likely because the microstructure of the polymer concrete was further densified with the increased addition amount of polymer binder, resulting in more enhanced water tightness. The hydrophobic characteristic of polyester binder could be

Table IV. Mixture Proportions of UP Polymer Concrete

PB ^a (wt %)	Filler (fly ash) (vol %)	Fine aggregate		Coarse aggregate (wt %)	Remarks
		River sand (wt %)	RCSS ^b (vol %)		
7.5	20	20	0	52.5	
		15	5		
		10	10		
		5	15		
		0	20		
8.0	20	20	0	52	
		15	5		
		10	10		
		5	15		
		0	20		
8.5	20	20	0	51.5	Shrinkage-reducing agent: PS ^e
		15	5		
		10	10		
		5	15		
		0	20		
9.0	20	20	0	51	
		15	5		
		10	10		
		5	15		
		0	20		

^aPB, polymer binder (UP 80% + PS 20%).

^bRCSS, rapid-cooled steel slag.

^cMEKPO, methyl ethyl ketone peroxide.

^dCoOc, cobalt octoate.

^ePS, polystyrene.

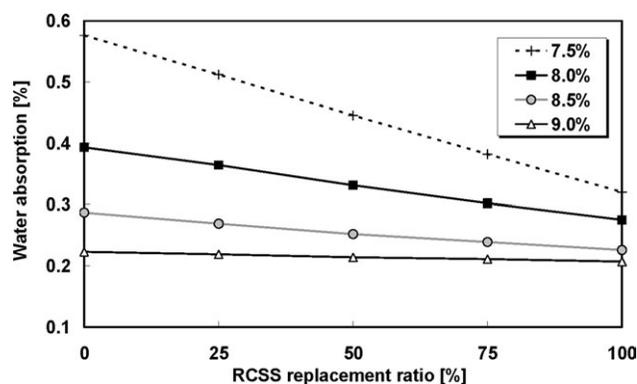


Figure 1. Water absorption versus replacement ratio of RCSS.

another probable reason that reduces the absorption rate with the increased binder amount. Moreover, the experimental result clearly shows that the absorption rate remarkably decreased as the RCSS replacement ratio increased. This is because the increased RCSS replacement ratio improved the mixture workability, which contributed to the better compaction of polymer concrete specimens. Finally, it appears that the significantly low absorption rate of the RCSS (i.e., 0.005%) compared to that of the river sand fine aggregate (i.e., 0.75%) contributed to the observed results as well.

Strengths and Hot Water Resistance

Figure 2(a) shows the variations of 28-day compressive strength depending on the replacement ratio of RCSS. As shown in the result, as the replacement ratio of RCSS increased, the compressive strength tended to increase until the polymer binder addition ratio reached up to 8.5%. However, it is interesting to note that the compressive strength decreased when the polymer binder addition ratio increased to 9.0%. This is because the high-polymer binder content promoted the fluidity and workability of the mixture, resulting in segregation of RCSS which has a high density. This finding indicates that the excessive use of polymer binder can be inefficient for compressive strength improvements of polymer concrete incorporating RCSS.

The variations of 28-day flexural strength depending on the replacement ratio of RCSS are shown in Figure 2(b). It is important to note that the flexural strength exhibited a different trend with the compressive strength. As the addition ratio of polymer binder and the replacement ratio of the RCSS increased, the flexural strength tended to keep increasing, whereas the compressive strength decreased when 9.0% polymer binder was added. This result occurred because the strength characteristics of polymer concrete are different depending on the failure modes; the previously published studies^{18–20} reported that polymer concrete is highly beneficial for enhancing the flexural and tensile strengths rather than the compressive strength because of its strong chemical bond characteristics between the aggregates and the polymer matrices. Their experimental results showed that the flexural and tensile strengths of polymer concrete were 5 and 6.7 times higher than those of conventional cement concrete, respectively, whereas the compressive strength was only 2.6 times higher than that of conventional one.

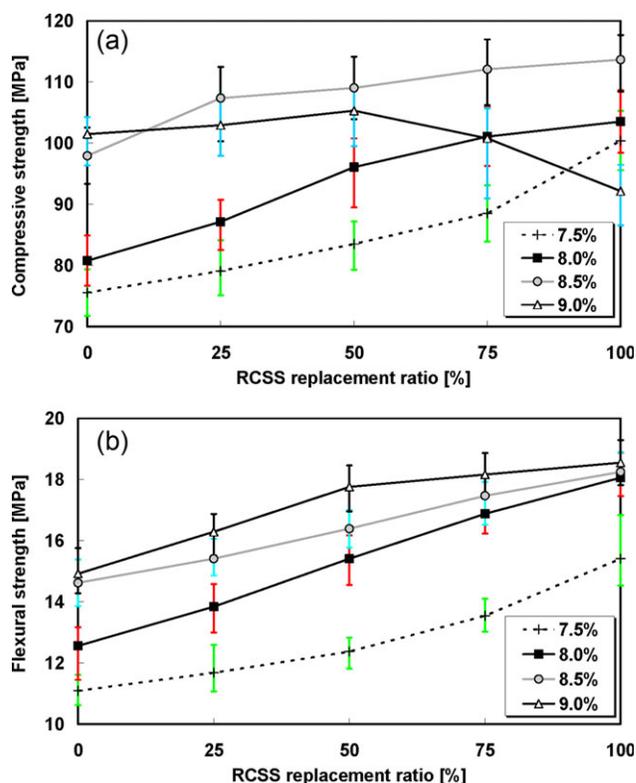


Figure 2. Strength versus replacement ratio of RCSS: (a) compressive strength and (b) flexural strength. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Figure 3(a) shows that the compressive strength reduction rates calculated based on the measured compressive strengths before and after the hot water resistance test. The result indicates that the compressive strength substantially decreased after the hot water resistance test. Overall, both polymer addition amounts of 7.5 and 8.0% showed the similar compressive strength reduction rates of around 30% regardless of the RCSS replacement ratio. However, the cases with the polymer binder addition amounts of 8.5 and 9.0% resulted in higher strength reduction rates of around 40%. Although the reduction rates generally increased as the replacement ratio of RCSS increased, the case incorporating 9.0% polymer binder displayed remarkably low-strength reduction rates as the RCSS replacement ratio became higher than 50%. This is because the absolute strengths measured before the hot water resistance test were quite low as shown in Figure 2(a).

Figure 3(b) shows the variations of flexural strength reduction rates depending on the amount of polymer binder and RCSS replacement ratio. The result shows that there was a significant loss in flexural strength after the hot water resistance test. It should be noted that the flexural strength yielded much greater strength reduction rates than the compressive strength. In addition, the result showed a different overall trend with the reduction rates of compressive strength; as the addition amount of polymer binder and the replacement ratio of RCSS increased, the flexural strength reduction rate almost proportionally increased.

The primary reason for this notable reduction in strengths after the hot water exposure is because the polymer binder

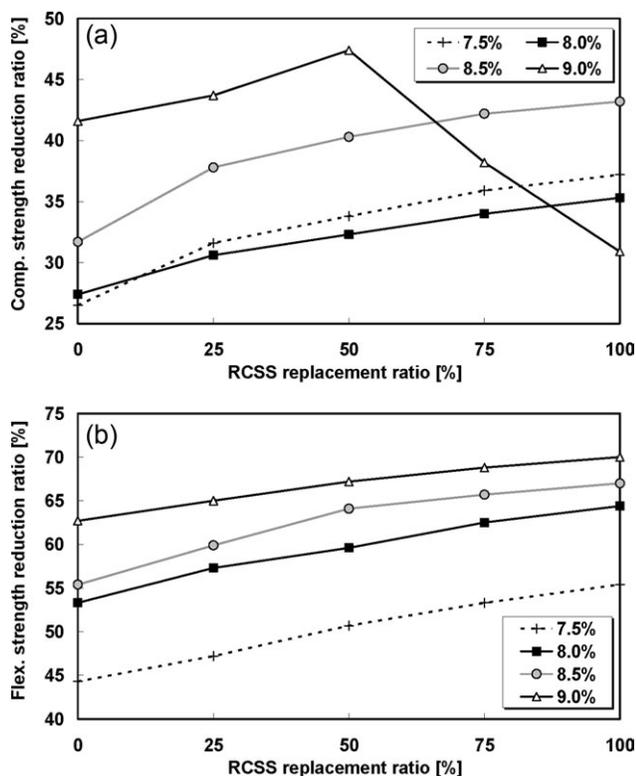


Figure 3. Strength reduction ratio after hot water resistance test versus replacement ratio of RCSS: (a) compressive strength and (b) flexural strength.

underwent degradation and decomposition under the elevated temperature condition and thus promoted the loss of bond strength among the components. Also, as the unsaturated polyester resin used in this study was basically a thermoset polymer, the brittleness of polymer concrete appreciably increased during the hot water test, which ultimately gave rise to the reduction in strengths. On the other hand, the strengths tended to decrease with the increased RCSS content. This is most likely because there was a large difference in coefficient of thermal expansion between the RCSS (i.e., $6 \mu\epsilon/^\circ\text{C}$) and the polymer binder (i.e., $50 \mu\epsilon/^\circ\text{C}$), causing microcracking in the vicinity of the RCSS particles after the hot water resistance test. Furthermore, as the thermal conductivity of the RCSS (i.e., 14.7 W/mK) is generally much higher than that of the unsaturated polymer binder (i.e., 1.7 W/mK), the polymer binder with a higher RCSS content was degraded more rapidly by hot water.

Pore Characteristics

Figure 4(a) shows the variations in total pore volume before and after the hot water resistance test. It is noted that as the replacement ratio of RCSS increased, the total pore volume tended to decrease. This trend resulted because, as previously described, the RCSS had about 55% less pore volume than the river sand and coarse aggregate. In addition, this finding is attributed to the fact that as the RCSS replacement ratio increases, the workability and fluidity of the mixture increase and thus form a denser microstructure with fewer air voids.

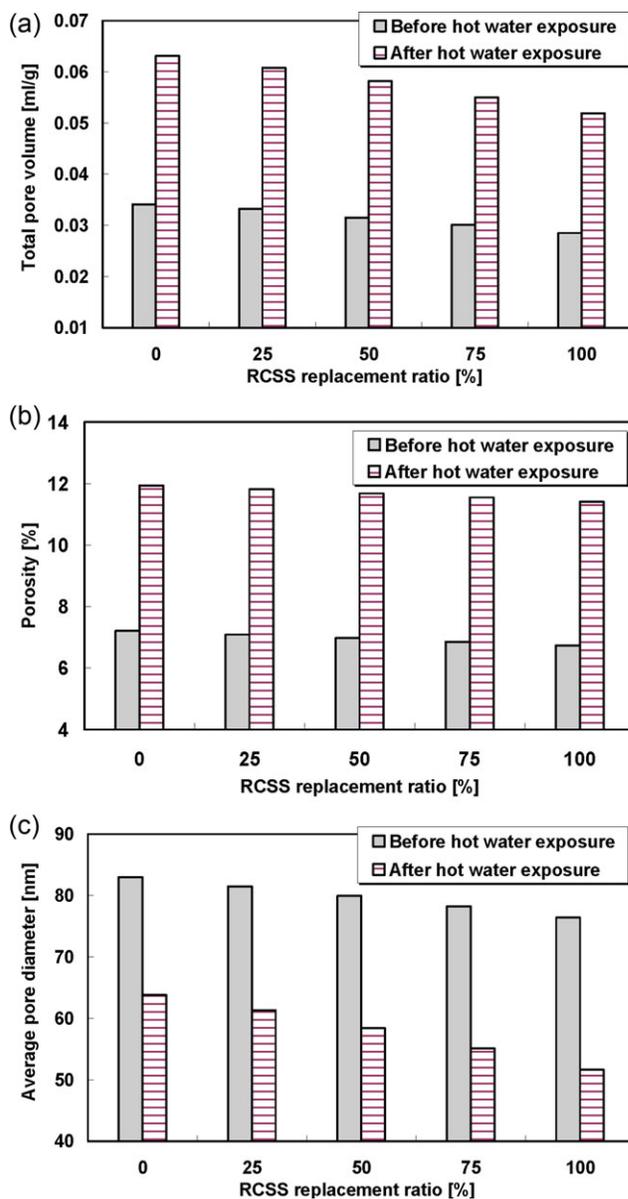


Figure 4. Pore characteristics versus replacement ratio of RCSS (polymer binder: 8.0%): (a) total pore volume, (b) porosity, and (c) average pore diameter. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

The substantial increase (i.e., approximately 85%) in total pore volume after the hot water resistance test is shown in Figure 4(a). This appears to be owing to the decomposition and degradation of polymer binder during the test at the elevated temperature as high as 90°C for 28 days. It should be also noted that, as shown in Figure 4(b), the result of porosity measurements showed a quite similar trend with that of the total pore volume measurements. This finding implies that the total pore volume is closely correlated with the porosity of a system.

The variations of average pore diameter before and after the hot water resistance test are shown in Figure 4(c) as a function of

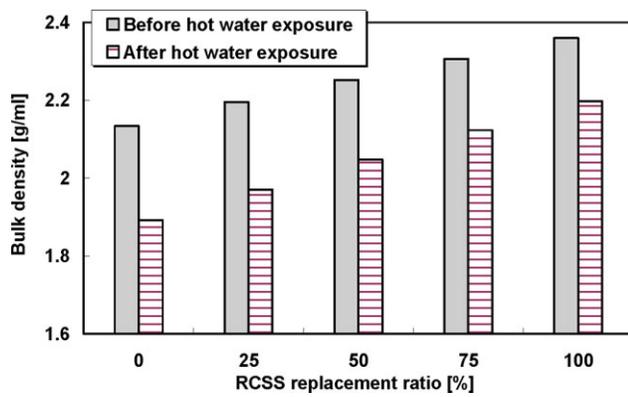


Figure 5. Bulk density versus replacement ratio of RCSS (polymer binder: 8.0%). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

RCSS replacement ratio. The result implies that as the replacement ratio of RCSS increased, the average pore diameter tended to decrease. It was also found that the average pore

diameter after the hot water resistance test reduced by around 30% compared to that measured before the test. The reason for the reduced average pore diameter after the hot water resistance test is most likely because the relatively larger pores were opened as the polymer binder was gradually decomposed and degraded under the elevated temperature condition.

Figure 5 shows the changes in bulk density before and after the hot water resistance test. The result indicates that the bulk density tended to increase as the replacement ratio of RCSS increased. This result was obtained because of the reason that the density of the RCSS—which has about 30% iron content—is much greater than that of the regular fine aggregate and coarse aggregate. The experimental result also revealed that the bulk density measured after the hot water resistance test was much smaller than that measured before the test. This finding also seems to be closely related to the degradation and decomposition of the polymer binder's microstructures during the hot water resistance test.

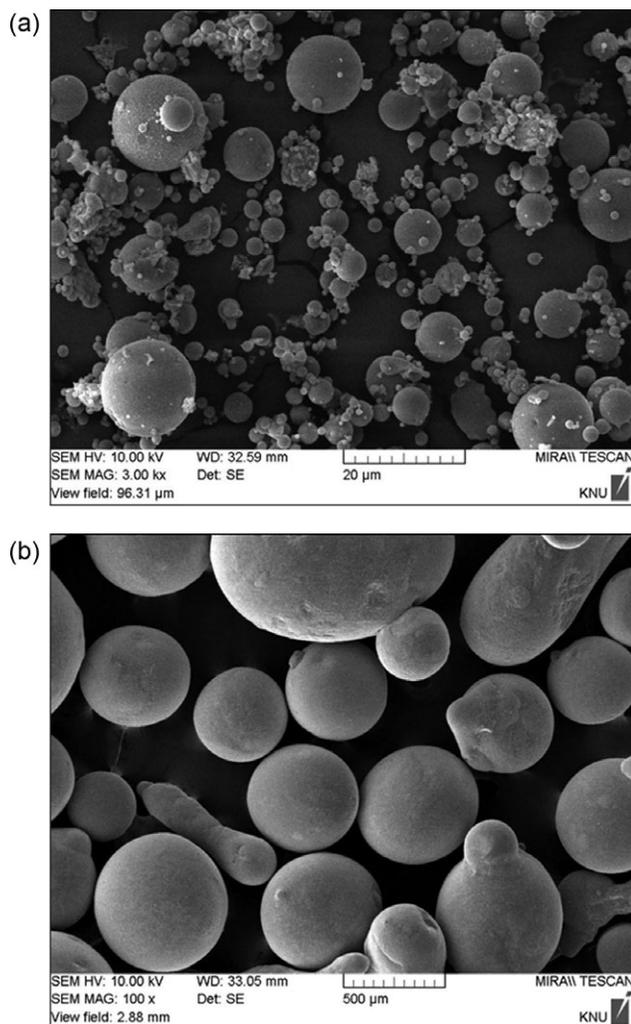


Figure 6. SEM micrographs of fly ash and RCSS: (a) fly ash and (b) RCSS.

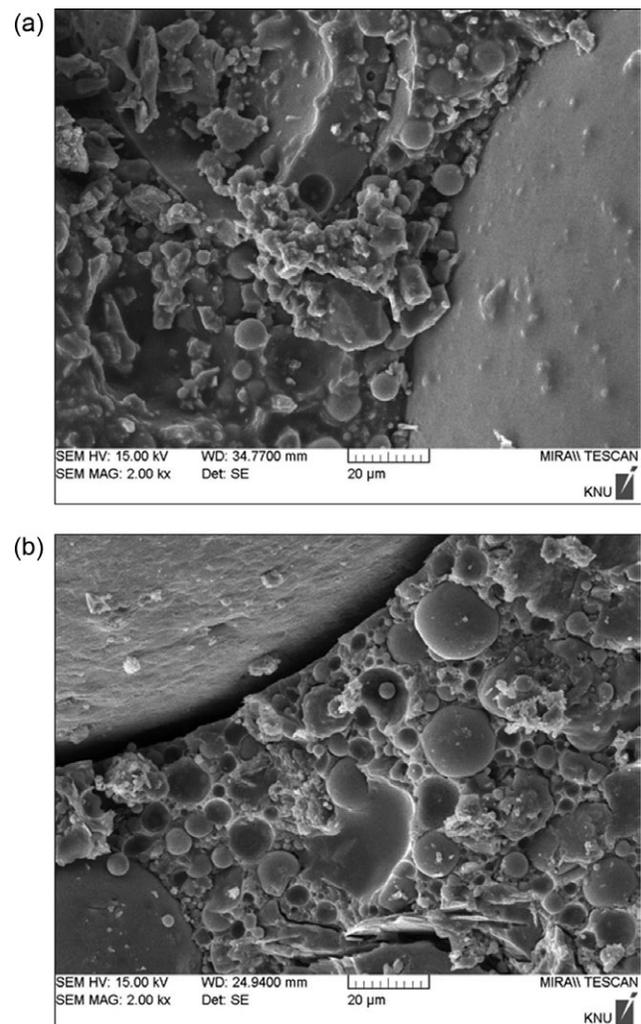


Figure 7. SEM micrographs of fracture surface before and after hot water resistance test (polymer binder: 8.0%): (a) before hot water resistance test and (b) after hot water resistance test.

Table V. Comparison of Production Costs Between Conventional and Developed Products (Price Unit: 1000 Korean won)

Materials	Price/ton	Price/m ³	Composition of conventional product (vol %)	Composition of developed product (vol %)	Price of conventional product (price/m ³)	Price of developed product (price/m ³)	Remarks (specific gravity)
UP resin	2300	2530	17.50	13.77	442.75	348.38	1.1
SRA ^a	2000	1900	5.06	3.99	96.14	75.81	0.95
Calcium carbonate	40	107	17.16	-	18.36	-	2.67
Fly ash	20	47	-	17.72	-	8.33	2.35
FA ^b	40	105	17.49	-	18.36	-	2.62
CA ^c	40	106	42.79	46.45	45.36	48.08	2.65
RCSS ^d	40	142	-	18.07	-	25.66	3.54
Additives (catalyst, accelerator)	-	-	-	-	16.17	12.73	3% of Polymer binder
Total price					637.14	518.99	

^aSRA, shrinkage-reducing agent.

^bFA, fine aggregate.

^cCA, coarse aggregate.

^dRCSS, rapid-cooled steel slag.

Observation of Microstructures with SEM

Figure 6(a,b) shows the micrographs of fly ash and RCSS observed with SEM, respectively. As shown in the Figure 6(a,b), the particle size of the fly ash was <20 μm, whereas that of the RCSS ranged from 0.1 to 1.2 mm. As the surfaces of the fly ash and RCSS are highly smooth and spherical, their surface areas are typically smaller than those of regular filler and fine aggregate. These smooth and spherical characteristics of the fly ash and RCSS also give rise to a ball-bearing effect, which ensures the enhanced workability during the mixing even with a small amount of polymer binder.

Figure 7 shows the micrographs of the fracture surface before and after the hot water resistance test. As shown in Figure 7(a), before the test, the polymer binder was completely fused to the surfaces of fly ash and RCSS, forming dense comatrix phases.^{21,22} After the test [Figure 7(b)], however, the polymer binder was severely decomposed and degraded, displaying clear boundaries among the polymer binder, fly ash, and RCSS. Particularly, large detachment cracking was observed at the interface between the polymer binder and the RCSS. This is because, as previously described, a significant discrepancy (i.e., about eight times) in coefficient of thermal expansion existed between the RCSS and the polymer binder.

Comparison of Material Costs Between Conventional and Developed Products

To evaluate the cost efficiency of the developed product, total material costs of the conventional and developed products were compared with each other as summarized in Table V. It is of great importance to note that the total material cost of the developed product was reduced by 18.5% compared to the conventional product. The decreased material cost primarily resulted from the reduction of polymer binder use by 21.3%, which occupies a substantial portion (i.e., about 85%) of the

entire material costs of polymer concrete. Based on the analysis results, it can be concluded that the use of spherical fly ash and RCSS in place of calcium carbonate filler and river sand can be an effective measure to reduce the total material costs, while improving the workability, strength characteristics, and durability of polymer concrete.

CONCLUSIONS

In this study, polymer concrete was fabricated using spherical RCSS and fly ash obtained as industrial by-products, and its performance was experimentally evaluated to investigate the potential applicability of RCSS and fly ash as alternative materials of fine aggregate (river sand) and filler (calcium carbonate). Furthermore, a cost efficiency analysis was performed by comparing the total manufacturing costs of the developed product with those of the conventional product. The findings of this study can be summarized as follows:

1. As the addition amount of polymer binder and the replacement ratio of RCSS increased, the absorption rate of polymer concrete tended to decrease. This is because the microstructure of the polymer concrete was further densified with the increased addition amounts of polymer binder and RCSS.
2. Overall, the 28-day compressive and flexural strengths showed an increasing trend with the increased addition amount of polymer binder and the replacement ratio of RCSS. However, in the case of 9.0% polymer binder addition and more than 50% RCSS replacement ratio, there was a sudden reduction in compressive strength. This result was obtained because the excessively increased workability of the mixture caused material segregation.
3. Significant losses in compressive and flexural strengths were found after the hot water resistance test. The strength reduction ratios were as high as 25–45% in the case of

compressive strength and 45–70% in the case of flexural strength.

4. The result of pore analysis showed that as the replacement ratio of RCSS increased, the total pore volume, porosity, and average pore diameter decreased, whereas the bulk density increased. However, after the hot water resistance test, the total pore volume and porosity appreciably increased, whereas the average pore diameter and bulk density were reduced.
5. The SEM observation revealed that polymer binder was severely decomposed and degraded after the hot water resistance test at 90°C. In particular, the polymer binder was highly segregated around the RCSS particles, indicating that the polymer concrete incorporating RCSS could be damaged under the elevated temperature condition.
6. By employing the spherical fly ash and RCSS obtained as industrial by-products in polymer concrete production, the use of polymer binder could be reduced by 21.3%, which in turn saved the total material costs by 18.5% when compared to the conventional product.

In this study, the experimental investigations were limited only to the polyester concrete specimens shortly after the hot water exposure. Upon further cooling and additional chemical shrinkage, microcracks may start to initiate and propagate perpendicular to the aggregate surface as the polymer matrix would undergo restrained volume reduction against the aggregates^{23,24}; this behavior may also significantly affect the strength and microstructure characteristics. Accordingly, further research needs to be performed in this regard.

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