

COMPRESSIVE STRENGTH AND NANOINDENTATION OF HIGH STRENGTH LIGHTWEIGHT MORTARS WITH WASTE GLASS BEADS

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ABSTRACT

In this study, the compressive strength and nanoindentation were analyzed by incorporating different types of waste glass beads (spherical (B) and crushed (CB)) as moisture carriers for internal curing to improve the mechanical properties of high-strength cementitious composites. The results showed that the compressive strength increased due to internal curing in the long term. Nanoindentation results showed an increase in density in both ITZ and paste regions. Therefore, it is believed that the potential of WGB has been demonstrated and can help improve the mechanical properties of cement composites. **Keywords:** waste glass bead, surface roughness, internal curing, compressive strength, nanoindentation

1. INTRODUCTION

The world's resources require disposal after use. Landfills and incineration are the most common methods of disposal. However, landfilling requires landfill sites and incineration produces carbon emissions, so there are concerns about environmental pollution. Among them, waste glass has been mainly landfilled for treatment after use. As a result, the amount of landfill space required for landfilling has increased continuously, and a solution to recycle it is needed. In the construction field, research has been conducted to improve the hydration of waste glass through internal curing as a lightweight aggregate. In addition, as a lightweight aggregate, it has the advantage of being lightweight as a substitute for general aggregates.

In concrete, proper curing is important to develop the mechanical and durability properties of concrete. Concretes with high levels of durability properties include high strength concrete (HSC), high performance concrete (HPC), and ultra high performance concrete (UHPC). However, due to the use of ultra-fine cementitious materials and low water to cement (W/C) ratio, high durability concrete may experience volume changes due to hydration and subsequent drying of the cement during curing[1]. To reduce shrinkage, external curing methods are often used to maintain a wet concrete surface[2]. However, external curing is difficult to apply to HSC with low W/C ratio, permeability, and porosity because moisture only acts on the surface layer of concrete and is not transferred to the interior[3,4]. In contrast, internal curing is performed to maintain the relative humidity (RH) inside the concrete. In this process, materials that can store moisture in the concrete are typically used, most commonly aggregates[3,4,5]. While the cement is hydrated, the moisture in the internal curing material is continuously released by capillary pressure due to the humidity difference between the

material and the matrix[5]. Suzuk et al. investigated the efficiency of internal wet curing using recycled waste porous ceramic coarse aggregate (PCCA) for concrete. When 40% PCCA was used with a W/B level as low as 0.15, non-shrinkage HPC was produced, which significantly increased the compressive strength[6]. Cuevas et al. studied the development of internal curing agent for concrete by adding expanded glass lightweight aggregate (EG-LWA) based on waste glass. They found that EG-LWA could effectively reduce autogenous shrinkage and suggested increasing the pore connectivity to use a higher percentage of saturated water content in future studies[7]. Yang et al. used paper sludge as a pore-forming agent to produce fine lightweight aggregate (LWA) from bauxite residue and investigated the internal hardening effect of LWA in high-strength mortar. The autogenous shrinkage was reduced by up to 88% because the fine and coarse pore structure of LWA made it easy for the internal curing water to drain out[8]. Existing studies have shown that the addition of moisture-containing porous aggregates to building materials based on ultrafine cementitious materials can significantly reduce self-shrinkage and drying shrinkage and have a positive effect on long-term strength development.

A previous study by Pyeon et al investigated the effectiveness of waste glass beads (WGB) as LWA to enhance the internal hydration of high strength cement composites (HSCC) and to understand the hydration process with age using thermodynamic modeling[9]. The WGBs used were of spherical (B) shape. The results of the study showed that with the addition of the same proportion of WGB, a difference in the volume of hydrate was observed under different curing conditions, indicating that the pre-wetted WGB released its water content, promoting the continuous supply of internal moisture during the curing period[9]. However, it is believed that the addition of WGB reduced the compressive strength of the matrix not only because of

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the pre-existing damage and poor mechanical properties, but also because the spherical shape and smooth surface of the WGB reduced the adhesion with the paste, resulting in a lower density in the Interfacial transition zone (ITZ) region [9]. As a result, the authors emphasized the need for further investigation to mitigate the effects on strength [9]. To improve this, it is necessary to consider the surface roughness of the WGB. Previous studies have shown that the rougher the surface of the WGB, the better the adhesion with the paste and the higher the density of the ITZ region, the better the compressive strength of the matrix compared to that of the spherical WGB [10]. Jia et al. investigated the mechanical properties of ITZ with the morphology of the coarse aggregate surface as a variable. The roughness of the aggregate affected the mechanical properties of ITZ and increased the bonding area of the aggregate-paste, explaining its effect on the mechanical bonding ability [11]. Hilal investigated the effect of aggregate roughness in lightweight aggregate concrete. When lightweight aggregate with a rough surface was used, the ITZ was strengthened, resulting in a 13.5% increase in compressive strength [12].

Therefore, in this study, the mechanical properties of WGB in high-strength cementitious composites were investigated as a function of surface roughness and replacement ratio. In addition, the properties of the paste-interfacial transition zone (ITZ)-WGB region were compared through nanoindentation measurements.

2. EXPERIMENTAL PLAN AND METHOD

2.1 Materials

The binders used in this study were ordinary portland cement (OPC), silica fume, and silica powder. The chemical composition of the binders used is summarized in Table 1. Silica fume and silica powder were used with a specific gravity of 2.2 and 2.5, respectively, and a surface area of 20 m²/g and 24 m²/g, respectively. As for the aggregate, silica sand was used to maintain the packing density [1]. Polycarboxylate superplasticizer for UHPC was used to ensure flowability, considering the minimum content that does not affect hydration, rather than to ensure the same slump flow of each mixture, considering that HSC usually has low W/C and W/B ratios and small particle size materials are used.

Table 1 Chemical composition of the binder

Chemical composition (%)	OPC	Silica fume	Silica powder
CaO	66.79	0.02	0.05
SiO ₂	16.14	99.00	99.00
Al ₂ O ₃	4.40	0.30	0.30
MgO	2.47	0.01	0.05
Na ₂ O	0.08	0.05	0.05
K ₂ O	1.18	0.02	0.05
SO ₃	3.23	0.05	0.05
Fe ₂ O ₃	3.83	0.05	0.05
TiO ₂	0.34	0.06	0.06

As an LWA, WGB is made from a waste glass powder material based on discarded sodium silicate. Since it is composed of sodium silicate-based glass, WGB is not affected by the alkali-silica reaction (ASR). The collected sodium silicate-based waste glass was ground into powder and then subjected to a special heat treatment that utilizes the thermal expansion of the Si structure of the glass [14]. The types of WGBs, B and CB, are manufactured in the same environment and are divided into B, which is formed into a sphere during the manufacturing process, and CB, which is formed into a sphere and then broken or not formed into a sphere [10].

Since WGB (B) has no surface friction, it has problems such as strength reduction and material segregation [9]. To solve this problem, we ground the WGB (B) and changed the surface roughness to acute and obtuse levels to increase surface friction. Surface texture is an important factor affecting absorption ratio in LWA [8]. Therefore, we obtained a spherical WGB (B) and a crushed WGB (CB) and compared the improvement in surface friction.

To obtain the absorption ratio and desorption ratio of WGB, 500 g of WGB (B, CB) was placed in each filter net in a completely wet state without weight change at a constant temperature and humidity. The weight of the water released was measured over time, and the desorption ratio was calculated by dividing the original weight by the weight of the released water. The absorption ratio is the total ratio minus the desorption ratio.

We used spherical WGB (B) with a particle size of 0.05-5.00 mm and crushed WGB (CB) with a particle size of 0.05-3.00 mm, as shown in Fig.1 and SEM images of WGB (B) and (CB) in Fig.2. The physical properties of WGB are: density of surface dry saturation is 1.00 g/cm³ and density in absolute dry state is 0.89 g/cm³. The water absorption of the WGB is 13.04% for B, 15.56% for CB, and 0.13% for the clay lump. This increased absorption ratio can be attributed to the presence of numerous internal voids in the beads. These findings highlight the differences in water absorption in cement composites. The higher water uptake of the crushed WGB suggests the possibility of enhanced internal hardening and density improvement, which is likely due to the difference in particle contact area and size between the two types. These results identify potential research directions to investigate different WGB applications in cement composites and promote a deeper understanding of internal hardening and performance enhancement. Recognizing these characteristics, it was decided to subject the WGBs to a 48-hour immersion process in 20 °C tap water. This immersion was performed under controlled conditions of 25 °C and RH of 55%, with fluctuations limited to within ± 5%. The immersion process continued until a weight stability state was achieved, ensuring WGB saturation. The water was then removed from the surface of the WGB and added to the matrix. This preconditioning step ensured that the internal voids of the beads were completely filled with water, allowing for a consistent and reliable subsequent experimental procedure.



Fig.1 Spherical WGB (B) and crushed WGB (CB)

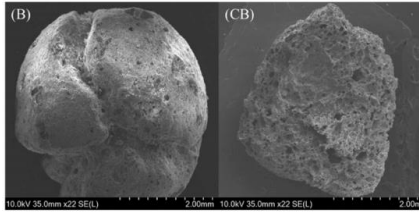


Fig.2 SEM images of WGB (B), (CB)

2.2 Mix Design

Table 2 details the mix ratios used to prepare the spherical (B) and crushed (CB) WGB specimens and the notation for each specimen. WGB was added to the mortar mix to investigate the effect of surface roughness. Two variants of WGB derived from recycled waste glass powder were included in this investigation: Spherical WGB (B) and crushed WGB (CB). These two types were explored to increase the specific surface area. To investigate the utility of oil palm shell (OPS) as a lightweight aggregate, silica sand replacement ratios of 10%, 30%, and 50% were selected to determine the applicability of WGB, based on the mixing ratios suggested by Aslam et al[15].

2.3 Experiment Preparation and Curing Conditions

First, the specimen preparation was a dry mix of binder material and silica sand. Once sufficiently mixed, water and superplasticizer were added and mixed for about 1.5 min. Once enough superplasticizer was released, spherical (B) or crushed (CB) WGB was added and further mixing was performed for approximately 1.5 min. Once mixing was complete, the fresh mixture was poured into a mold for ASTM C230/C230M-21 and several tests were performed. After 24 hours of curing at room temperature (RT) ($20 \pm 2^\circ\text{C}$, $55 \pm 5\%$), the samples were extracted from the molds and steam cured ($80 \pm 2^\circ\text{C}$, $90 \pm 5\%$) for 72 hours. The specimens were then sealed with aluminum tape and curing continued at RT

Table 2 Mix proportions (mass ratio) of the specimens

Mixture ID	Cement	Silica fume	Silica powder	Water	SP admixture	Sand	WGB (B)	WGB (CB)
P						1.0	-	-
B10						0.9	0.1	-
B30						0.7	0.3	-
B50	1	0.26	0.30	0.30	0.03	0.5	0.5	-
CB10						0.9	-	0.1
CB30						0.7	-	0.3
CB50						0.5	-	0.5

until 180 days. These curing conditions were chosen because they can enhance strength development and durability during the early curing stage of high-strength mortars exposed to high temperature or high pressure[16]. The selected conditions promote strength development and improve durability, which benefits overall performance.

2.4 Experimental Method

The compressive strength of the specimens mixed with WGB was measured at 1, 3, 7, 28, and 180 days according to ASTM C109/C109a.

Nanoindentation measurements were performed to determine the elastic modulus and plastic hardness of each region where the cement paste-ITZ-WGB was present. The modulus of elasticity describes the stiffness of a material, while plastic hardness describes its resistance to permanent deformation and damage. Thus, it is possible to evaluate the microstructural properties of the matrix as a function of the volume content and type of WGB admixture. The degree of polishing of the surface to be measured is the most important factor[15,16]. To ensure the reproducibility of the surface to be measured and to maintain uniform penetration of the indenter, the top surface of the $20 \times 20 \times 20$ mm cube specimen prepared for the nanoindentation test was polished to 1000 and 1500 mesh. The final polishing was performed at 3000 mesh to ensure that a smooth nanoscale surface area was in contact with the nanoindentation probe. Measurements were then taken using a microscope after the paste-ITZ-WGB on the surface was clearly distinguishable.

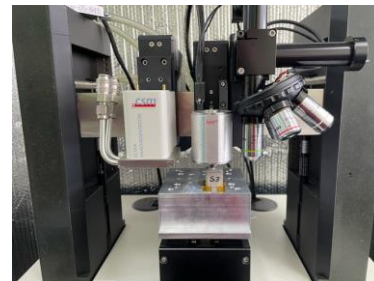


Fig.3 Nanoindentation tester

Nanoindentation was performed with the NHT2, a nanohardness tester from the Korea Atomic Energy Research Institute, using a standard Berkovich probe. The indentation was held for 10 seconds for load control under a peak force of $100 \mu\text{N}$ to $5000 \mu\text{N}$. The load was increased at a constant rate of 10 mN/min until it reached its maximum value. After 10 seconds at the maximum

level, the load was removed at the same rate until the material was no longer in contact with the indenter tip. The load depth curve of the trapezoidal load history was obtained with the same condition by referring to a previous study[17]. Fig.3 shows the testing machine for the nanoindentation test. This testing machine was used to investigate the properties of the ITZ interface comprising the paste-ITZ-WGB to determine the relationship between the morphology of the microstructure and the reaction kinetics of the cement[19].

3. RESULTS AND DISCUSION

3.1 Compressive Strength

Fig.4 shows the compressive strength of the HSC for each WGB type and the fracture pattern of each specimen. The strength development of the specimens differed depending on the WGB type. In Group B, the failure of the LWA occurred earlier than the matrix due to its lower strength. This trend became more evident as the percentage of WGB content increased, indicating that the efficiency of matrix strength decreases as the volume of LWA increases[18]. What is noteworthy about these results is that although the decrease in LWA strength appears to be an important factor, an internal curing effect can be expected given the rate of strength increase. The lower rate of strength reduction in the CB test group compared to the B test group is likely due to the higher efficiency of matrix strength due to the irregular surface roughness, specific surface area, and small particle size of the aggregate. Long-term strength was improved when WGB was used. The reason for this was the high interfacial bonding with the cement paste due to the absorption properties of WGB. The mechanism of interfacial bonding (ITZ) between WGB and cement paste is as follows: (1) The porosity and coarse granularity of WGB are favorable for the growth of some hydration products on the surface. (2) The water released from the WGB promotes hydration and optimizes the ITZ structure[21]. Due to the aforementioned effects, WGB facilitates the surface adhesion of the slurry during mixing. In addition, group CB has a coarser granularity, which results in a higher surface roughness and thus forms a stronger ITZ, which

improves the overall system strength.

From a long-term strength perspective, Group B showed a 40% strength improvement between 28 and 180 days. When comparing strength gains between Group CB and Group B, Group CB showed higher gains in initial strength recovery. Group B showed lower initial strength gains, but higher strength gains due to the gradual increase in the bond between WGB and cement paste. From a long-term strength perspective, Group B showed a 40% strength improvement between 28 and 180 days. When comparing strength gains between Group CB and Group B, Group CB showed higher gains in initial strength recovery. Group B showed lower initial strength gains, but higher strength gains due to the gradual increase in the bond between WGB and cement paste.

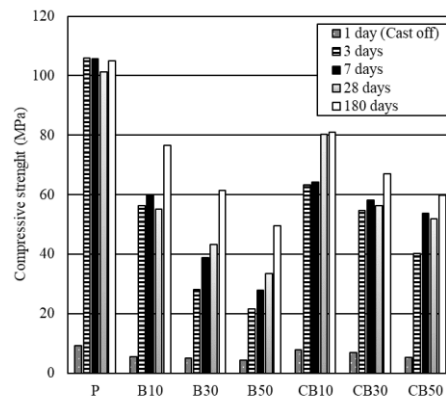


Fig.4 Compressive strength of each specimen

Therefore, we can see that the WGB particle shape has a significant relationship with the bonding force of the cement paste. If the goal is to improve the strength while keeping the initial binding force high, coarse-grained WGB (CB) can be considered advantageous. In general, the strength development varied as the proportion of matrix material in the specimen was varied. As the WGB content increases, the content of the matrix decreases; However, internal hardening can occur as moisture trapped in the WGB is released into the matrix, increasing the hydration potential and the rate of strength development.

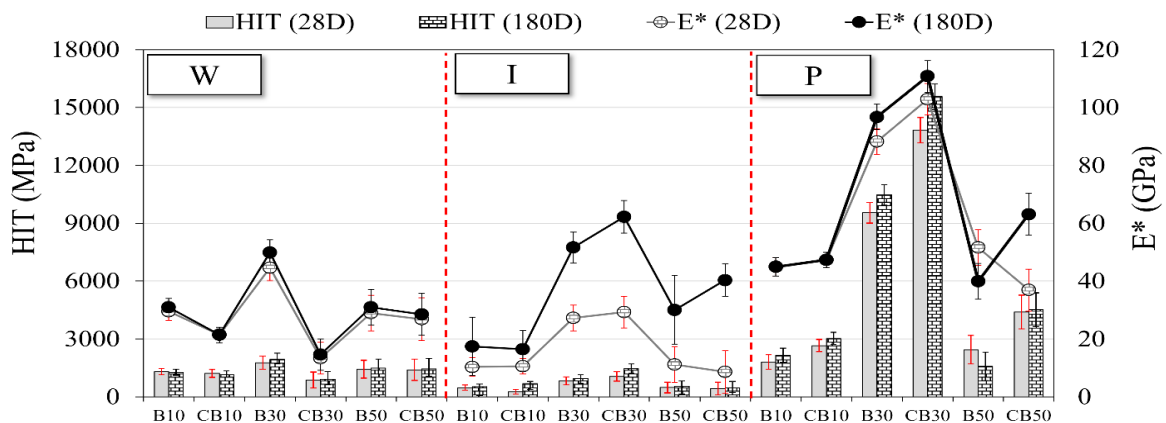


Fig.5 Nanoindentation of each specimen

3.2 Nanoindentation

Nanoindentation has been used to determine the mechanical properties of various aggregates and pastes and can be related to the microscopic nature of the materials used in the specimen[22]. Fig.5 shows the elastic modulus (E^*) and plastic hardness (HIT) of cement paste-ITZ-WGB cross-sections with nanoindentation as a function of age. In Fig.6, the nanoindentation measurement area is shown on the specimen surface with the SEM imaging. The differences in the mechanical properties of cement paste, ITZ, and aggregate were analyzed, and the E^* and HIT values were obtained for each cross section. We also averaged these values for each cross-section of the whole specimen. The modulus of elasticity and plastic hardness are different for each cross section. Previous studies have suggested that these differences are due to the degree of hydration and changes in the structure of the hydration products. The unique elastic properties of C-S-H present in the paste and ITZ may affect the elastic modulus of those sections[23].

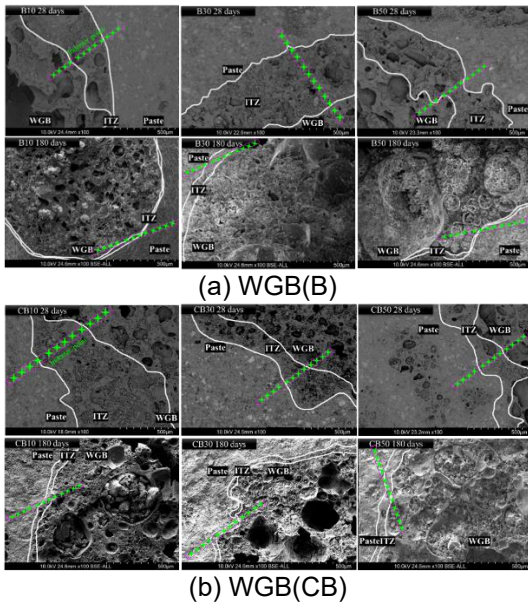


Fig.6 The area of nanoindentation measurement marked with scanning electron microscopy (SEM) imaging

Although similar elastic modulus and plastic hardness values were measured in the WGB sections, each specimen showed different indentation hardness and elastic modulus values in the ITZ-paste sections depending on the WGB type. The E^* and HIT values of the ITZ and paste sections of the B30 and CB30 specimens were higher than the other specimens, which is consistent with the results of the analysis in Section 3.1, which considered macroscopic properties. It was also concluded that these higher measurements were due to the increased volume of the closed pores identified by SEM analysis and the formation of hydrates in the pores of the WGB.

In the paste sections, the micromechanical properties were higher for CB30 than for B30. In other words, the rougher surface CB type improved the

mechanical properties more than the smoother B type. In addition, regardless of the roughness, higher WGB replacement ratios were obtained at 30%, indicating that these specimens had adequate distribution of water-saturated WGB, which improved the internal curing by water desorption. Previous studies have shown that the hardness decreases and gradually stabilizes with increasing distance from the LWA surface due to the difference in the degree of hydration between the ITZ and the paste[24]. For a water-saturated LWA, both the degree of hydration and the density of the paste around the ITZ are significantly enhanced as the water desorbs[25]. In other words, porous materials such as WGB may degrade the strength of HSCs and HPCs, but may be favorable for internal curing.

Thus, the nanoindentation measurements improved over time for WGB replacement ratios of about 30-50%, indicating that the water trapped in the WGB gradually hardened the ITZ through continuous hydration with the surrounding cement particles, contributing to the overall strength improvement.

4. CONCLUSIONS

In this study, the mechanical properties of WGB in high-strength cementitious composites as a function of surface roughness and replacement ratio were investigated. The main contributions can be summarized as follows.

- (1) The strength of HSCs decreased as the percentage of WGB content increased. However, this strength change varied from specimen to specimen depending on the surface roughness of the WGB. The addition of crushed WGB (CB) resulted in a higher efficiency of matrix strength due to its irregular surface roughness and small particle size. In addition, the higher the desorption point, the higher the strength compared to the results of WGB adsorption experiments.
- (2) Comparing the B and CB specimens, significant improvements were observed when the replacement ratio of WGB was 30% and 50%. This improvement can be attributed to the change in ITZ hydration between the WGB and the paste. This process involves the release of moisture previously trapped within the WGB, resulting in increased density in both the ITZ and the paste due to moisture interaction with the cement particles.

The addition of lightweight aggregates inevitably reduces the compressive strength of mortar and concrete. Nanoindentation measurements confirm that the elastic modulus and plastic hardness of ITZ increase due to the internal curing effect of WGB. This can partially mitigate the decrease in compressive strength caused by lightweight aggregates. In this study, 30% WGB incorporation is considered suitable. Further studies should consider the appropriate proportion of WGB in normal aggregate and curing environment to expand the applicability of WGB. This is expected to increase the applicability of WGB.

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