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Study on the Effect and Mechanism of Alkali–Silica Reaction Expansion in Glass Concrete

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Abstract: The suppression of ASR expansion hazards of glass concrete has always been a key and hot issue in the research of glass concrete. According to the ASTM C1260-14 fast mortar rod method, glass sand and glass powder act as fine aggregate and auxiliary cementing material, respectively. The changes in expansion rate with different amounts of glass sand content and different particle sizes of glass powder in mortar rods were compared, and the effects of glass sand content and the glass powder particle size on the expansion of ASR were analyzed. SEM was used to compare and analyze the microstructure of mortar rods to explore the mechanism of ASR expansion of glass concrete, and the results showed that the addition of glass powder had a certain inhibitory effect on ASR expansion. The larger the particle size of glass powder was, the better the inhibition effect on ASR expansion and the longer its duration. Compared with the three groups of experiments of 0–13 μ m, 13–38 μ m, and 38–75 μ m, it was found that the influence of the glass powder particle size on the expansion of ASR was weaker than that of dosage. The inhibitory effect of glass powder on ASR expansion is related to the fact that glass powder is more involved in pozzolanic reaction in the early hydration process.

Keywords: glass; concrete; ASR expansion; CSH gel; mechanism



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1. Introduction

With the continuous development of industrialization and urbanization in various countries, glass is widely used in various fields of production and life but is followed by a sharp increase in waste glass. The production of waste glass in the world has been increasing at an astonishing rate nowadays, becoming an important source of environmental pollution. However, using waste glass in building materials is a more effective way of making use of waste glass, which can reduce solid waste and save building material resources.

At present, the research on waste glass concrete, on both national and international levels, is mainly divided into the following parts: In the first group of studies, the ground glass is used as concrete fine aggregate [1–5], while in the second, it is used as inert filling material in concrete [6,7]; the third theme of research is to produce cement as cement clinker, and in the fourth, cement is partially replaced as an auxiliary cementitious material [8–10]. It is found that when waste glass is used as aggregate, the mechanical properties of concrete are reduced, mainly due to the existence of a weak interface [11,12]. In addition, an alkaliaggregate reaction will occur when the aggregate particles are larger (mainly refers to the particle size larger than 1.2~1.5 mm), which leads to the expansion failure of concrete by water absorption [11,12]. This phenomenon does not occur when the glass is ground into powder, but the potentially high alkali content caused by it must be noted. Glass has a pozzolanic activity after being ground into powder, which is also a much debated problem among researchers [13–15].

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At first, a large number of researchers mainly devoted themselves to the study of waste glass in concrete aggregate for a long time [3–5]. Due to the alkali in cement and a large amount of active silica in glass, which leads to considerable alkali-aggregate reaction and expansion damage of concrete [10] and thus has made the research stagnant. Although researchers have proposed several methods to reduce ASR expansion, such as replacing the original cement with pozzolanic cement, adding fiber materials to waste glass concrete, adding lithium compounds, etc. [16], the outcomeis still not satisfactory. Some researchers also used glass as the raw material for the production of cement. However, the experiments showed that the increase in alkali content in cement after the addition of glass addition easily causesan alkali-aggregate reaction, which is not beneficial to the performance of concrete [17]. Mohajerani et al. proved that when the glass powder is ground to a certain extent, it does not cause ASR, and it also has pozzolanic activity, which can improve the performance of concrete [16]. Researchers found that when the particle size of glass powder is less than 300 μm , it shows significant pozzolanic activity, and the ASR reaction will become very weak [18]. Meyer et al. found that when the cement replacement rate is low and the age is 90 days, the pozzolanic activity of glass powder is higher than that of fly ash when the size is less than 100 µm [19]. Jani et al. proved that glass powder has good pozzolanic activity when the particle size is less than 20µm. The activity indices of 7d, 28d, and 90d reach 82%, 95%, and 102% [20], respectively. Schwarz et al. found that when the waste glass powder is added into concrete, the compressive strength of concrete is the best, and the hydration effect is enhanced when the content is 10% [21].

However, an earlier study found [22–24] that glass concrete was prone to alkali–silicic acid reaction (ASR), resulting in local expansion and cracking of the concrete, which severelyaffected the safety of concrete structures. This is the most important factor restricting the development and application of waste glass concrete. Therefore, the suppression of the ASR expansion hazards of glass concrete has always been a key and hot issue in glass concrete research. At present, the main measures to inhibit the expansion of glass concrete ASR include the mixing of fly ash, metakaolin, slag, silica fume, and other mineral admixtures [25–29], mixing chemical admixtures such as lithium-containing compounds and aluminum-containing compounds [30–34], controlling the size of glass particles [35–38], etc. Interestingly, many scholars have shown [39–41] that ground glass powder itself is an effective ASR expansion inhibitor. As an auxiliary cementitious material for concrete, glass grinding can stimulate the activity of volcanic ash, helping to improve the mechanical properties and durability of concrete and reduce the ASR hazards of glass concrete.

In this paper, according to ASTM C1260-14 "Standard Test Method for Alkali Potential Reactivity of Aggregate. Cement Stick Method", the ASTM C1260 rapid mortar bar method was adopted [42]. The glass sand and glass powder prepared by waste glass were used as aggregate and auxiliary cementitious materials, respectively. The river sand and cement were replaced by equal mass, the expansion rate of mortar rods with a different glass sand content and different particle size of glass powder was compared, and the effects of glass sand content and particle size of glass powder on ASR expansion were analyzed. The microstructure of mortar bars was analyzed by scanning electron microscopy (SEM), to further explore the mechanism of glass sand content and glass powder particle size on ASR expansion of glass concrete. The inhibition mechanism of glass powder on ASR expansion may be due to the potential pozzolanic property of ground glass powder. Glass powder is mixed into concrete as auxiliary cementitious material to provide relevant theoretical support for the recycling and reuse of waste glass in concrete.

2. Experiment

2.1. Raw Materials

The experimental cement is P.O. 42.5 ordinary Portland cement produced by Guilin Conch cement Factory, its specific surface area is $420.9~\text{m}^2/\text{Kg}$, density is $3150~\text{kg/m}^3$. The main chemical composition is shown in Table 1. The sand used in the experiment is the Lijiang River sand collected locally in Guilin; its apparent density is $2630~\text{kg/m}^3$. ASTM

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C1260-01 "Standard Test Method for Alkali Potential Reactivity of Aggregate. Mortar Stick Method" and TB/T 2922.5-2010 "Rapid Mortar Rod Method for Testing Alkaline Reactivity of Aggregate for Railway Concrete" were screened, to obtain the river sand particles with different grain sizes, and then the experimental sand was prepared according to the gradation shown in the specification. The glass was made of brown beer bottles recycled from the local waste station in Guilin. NaOH using alkali with purity \geq 98% was purchased from Tiantian instrument and equipment Co., Ltd. (Guilin, China).

| Table 1. Chemical compositions of cement and glass (wt. ' | ons of cement and glass (wt. % | o). |
|--|--------------------------------|-----|
|--|--------------------------------|-----|

| Compositions | SiO_2 | Al_2O_3 | CaO | Na ₂ O | K ₂ O | Fe_2O_3 | MgO | SO_3 | TiO ₂ | Other |
|--------------|---------|-----------|------|-------------------|------------------|-----------|------|--------|------------------|-------|
| Cement | 20.2 | 4.7 | 61.9 | 0.19 | 0.82 | 3.0 | 2.6 | 3.9 | / | 1.9 |
| Glass power | 72.2 | 2.55 | 7.11 | 0.42 | 0.64 | 2.47 | 0.46 | / | 0.15 | 1.03 |

2.2. Preparation of Glass Powder with Different Particle Sizes

The initial diameter range of glass sand from waste glass products purchased from a waste recycling station was mainly 233–282 μm . The specific surface area was 365.7 m^2/kg , and the density was 2470 kg/m^3 . The main chemical components are shown in Table 1. The final particle size of glass powder was 0–13 μm , 13–38 μm , and 38–75 μm by cleaning, crushing, ball milling, screening, and drying. The preparation process is shown in Figure 1. The specific surface area and density of each particle size of glass powder are shown in Table 2.



Figure 1. Flowcharts for preparation of glass powder with different particle sizes. (a) Cleaning; (b) Preliminary crushing; (c) Jaw crushing; (d) Ball-milledinto powder; (e) Dissolving into water; (f) Screening by Wet sieve method; (g) Drying; (h) Secondary Ball-milled.

Table 2. Blaine surface area and density of glass power with different particle sizes.

| Particle Size (μm) | Blaine Surface Area (m²/kg) | Density (kg/m³) |
|--------------------|-----------------------------|-----------------|
| 0–13 | 729.0 | 2510 |
| 13–38 | 521.8 | 2490 |
| 38–75 | 441.2 | 2480 |

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2.3. Experimental Method

Glass powder and glass sand were used as auxiliary cementing material and aggregate, respectively, to replace cement and river sand with equal mass. According to the mixture ratio shown in Table 3, mortar bar specimens were made, and the specimen size was $25 \text{ mm} \times 25 \text{ mm} \times 280 \text{ mm}$. A total of 12 sets of mortar rod specimens were made in this experiment, with 3 specimens in each group. Among them, JZ0, JZ20, and JZ100 were the control specimens without glass powder and glass sand content (0%, 20%, 100%). The molds of each group were removed after pouring for 24 h. The initial length of the mortar bar was measured at room temperature and then placed in a 1.0 mol/L NaOH curing solution in a constant temperature water bath. The temperature was maintained at 80 °C \pm 1. The lengths of mortar rods at 14d were tested. The specimen was taken out of the thermostat, and the length test of the mortar bar was completed within 15 s to reduce the influence of ambient temperature on the test results. The length of each mortar bar at different ages was measured, and the expansion rate of each specimen at different ages was calculated by Equation (1); then, the final expansion rate of each group was obtained. The microstructure and reaction product composition of specimens after curing for 14 d were observed by scanning electron microscopy (SEM), and the effect and mechanism of ASR expansion of glass concrete were analyzed.

$$\varepsilon_t = \frac{L_t - L_0}{L_0 - 2\Delta} \times 100 \% \tag{1}$$

where ε_t is the expansion rate of the test piece on day t; L_t is the length of the test piece on day t; L_0 is the initial length of the test piece; Δ is the length of the test probe.

| Number | Glass Powder Particle Size /µm | Glasspowdercontent /wt.% | Glass Sand Content /wt.% | Cement /g | Glass Powder/g | Glass Sand /g | Sand /g | Water /mL |
|--------|--------------------------------------|-----------------------------|--------------------------------|--------------|-------------------|---------------------|------------|--------------|
| JZ0 | / | 0 | 0 | 400 | 0 | 0 | 900 | 188 |
| GP1 | 0~13 | 20 | 0 | 320 | 80 | 0 | 900 | 188 |
| GP2 | 13~38 | 20 | 0 | 320 | 80 | 0 | 900 | 188 |
| GP3 | 38~75 | 20 | 0 | 320 | 80 | 0 | 900 | 188 |
| JZ20 | / | 0 | 20 | 400 | 0 | 180 | 720 | 188 |
| GP4 | 0~13 | 20 | 20 | 320 | 80 | 180 | 720 | 188 |
| GP5 | 13~38 | 20 | 20 | 320 | 80 | 180 | 720 | 188 |
| GP6 | 38~75 | 20 | 20 | 320 | 80 | 180 | 720 | 188 |
| JZ100 | / | 0 | 100 | 400 | 0 | 900 | 0 | 188 |
| GP7 | 0~13 | 20 | 100 | 320 | 80 | 900 | 0 | 188 |
| GP8 | 13~38 | 20 | 100 | 320 | 80 | 900 | 0 | 188 |
| GP9 | 38~75 | 20 | 100 | 320 | 80 | 900 | 0 | 188 |

Table 3. Mix proportion of the sample.

3. Results and Analysis

3.1. Analysis of Potential ASR Hazards of Aggregate

The 14-day expansion rate of mortar bars of each group is shown in Figure 2. It can be seen from Figure 1 that when glass sand replaces river sand by 20% and 100%, the 14-day expansion rate of concrete is 0.179% and 0.212%, respectively, which is much larger than the critical value of less than 0.1% required by the code [29]. This shows that there are great safety risks in directly applying waste glass to concrete in the form of glass sand. The necessary ASR suppression treatment must be carried out. Further study found that the expansion rate of concrete mixed with glass powder decreased significantly. For example, the 14-day expansion rate of the sample with a glass powder content of 20% and a particle size of 13–38 μ m was only 0.02%, which was much lower than the 0.1% required by the specification, which indicated that the glass powder could effectively restrain the ASR of concrete, reduce the ASR expansion rate, and improve the service performance of concrete.

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At the same time, when the glass powder content is 20%, and the glass sand content is 100%, the expansion rate of the 14 days concrete is still more than 0.1%. For example, the expansion rate of the sample with a particle size of 13 μ m–38 μ m is 0.129%. Although the expansion rate of the sample without glass powder (0.212%) has decreased significantly, there is still a risk of ASR expansion during the service of glass concrete. However, the production cost of glass powder is much higher than that of glass sand, so the application of waste glass in concrete should be combined with glass powder and glass sand according to a certain proportion to achieve the purpose of complementary advantages.

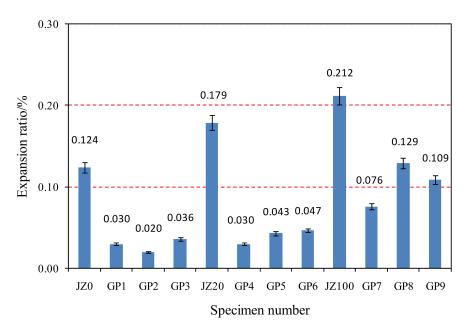


Figure 2. Histogram of 14-day-old expansion rate of mortar rods in each group.

3.2. SEM Image Analysis of Glass Concrete

Figure 3 is the SEM image of the specimens with different glass sand content without glass powder during the curing age of 14 days. It can be clearly seen that with the increase of glass sand content, more fluffy and thin-skinned reaction products are generated on the particle surface, and the overall structure is loose and there are many gaps. Based on the appearance characteristics of the reaction product and the energy spectrum analysis of the thin-skinned reaction product in zone A, it is inferred that the reaction product is ASR gel. It shows that due to the addition of glass sand, a considerablealkali–silicic acid reaction (ASR) occurred in the composite cementitious material system, resulting in a large amount of ASR gel. These ASR gels will absorb a lot of water and increase their volume, leading to a visible increase in the expansion rate of the sample, shown macroscopically. It is proved that severe ASR occurred in mortar rods without glass powder, and a large number of ASR gels are produced, which will further demonstrate the potential ASR hazard of local river sand in Guilin from the microscopic point of view [24].

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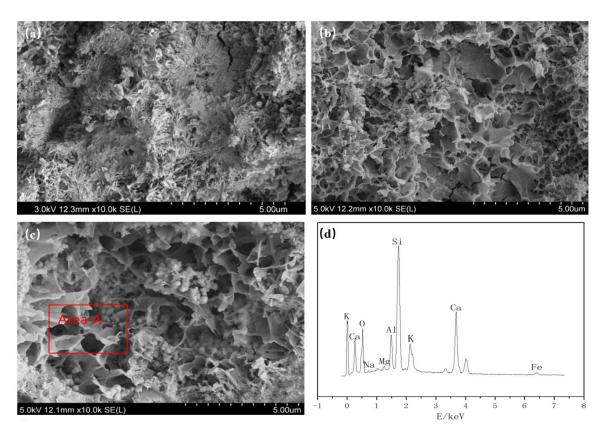


Figure 3. SEM images of 14dspecimens with different amounts of glass sand content, without glass powder, along with the energy spectrum analysis: (a) JZ0 (glass sand content: 0%); (b) JZ1 (glass sand content: 20%); (c) JZ2 (glass sand content: 100%); (d) energy spectrum analysis of area A.

Figure 4 shows the SEM images of specimens with 20% glass powder and 13–38 μm glass powder with different glass sand content for 14 days. It can be seen from Figure 4a that after adding glass powder, a large number of fibrous reaction products are formed on the surface of the concrete particles. These reaction products are closely packed and overlap each other to form a dense structure without a characteristic morphology. It can be judged as CSH gel by the power classification. It is inferred that the glass powder with a smaller particle size can stimulate its potential pozzolanic activity. The composite cementitious material is mainly a denser CSH gel formed by a volcanic ash reaction, which leads to a denser concrete structure and smaller voids. The macroscopic characterization is that the expansion rate of the specimen decreases. At the same time, by comparing the SEM images of GP2/GP5/GP8 and the control group JZ0/JZ1/JZ2, it can be found that when the concrete mix ratio is the same, the amount of loose ASR gel generated on the surface of the GP2/GP5/GP8 concrete particles mixed with glass powder is less, compared with JZ0/JZ1/JZ2 without glass powder, and the surface voids are relatively reduced. It is characterized from the microscopic level that the ground glass powder can effectively inhibit the ASR expansion of the mortar rod specimens. Through the comparison, it is not difficult to find that with the increase in he amount of glass powder, the loose structure and "paper-like" ASR gel gradually decreases, and the microstructure of the mortar rod becomes increasingly denser, indicating that the reaction products are more CSH gel. The inference in the above experiments is further verified, that is, "the increase in glass powder content will enhance the inhibitory effect on ASR expansion" [43].

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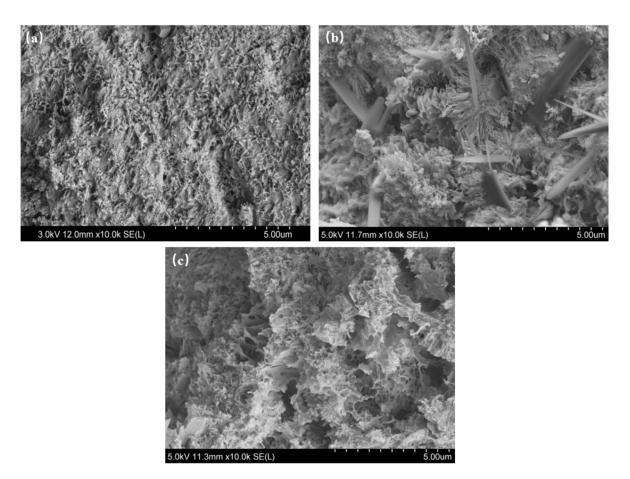


Figure 4. SEM image of different glass sand content specimens with 20% content and 13–38 μm glass power particle size (14d): (a) GP2 (glass sand content: 0%); (b) GP5(glass sand content: 20%); (c) GP8(glass sand content: 100%).

3.3. Analysis of the Expansion Mechanism of Glass Concrete ASR

When the concrete contains reactive aggregate, both favorable pozzolanic reaction and harmful alkali–silica reaction (ASR) may occur. When a pozzolanic reaction occurs in concrete, dense CSH gel is generated, and the reaction formula is shown in Formula (2). When ASR occurs in concrete, loose ASR gel is generated. The ASR reaction formula is similar to the pozzolan reaction formula, but Na⁺ and K⁺ are replaced by Ca²⁺. The reaction formula is shown in Formula (3). The two reactions coexist and compete with each other.

$$SiO_2 + Ca^{2+} + 2OH^- \rightarrow n_1 CaO \cdot SiO_2 \cdot n_2 H_2 O$$
 (2)

$$SiO_2 + 2Na^+(K^+) + 2OH^- \to Na_2(K_2) \cdot SiO_3 \cdot H_2O$$
 (3)

Combined with the change rule of expansion rate and the comparative analysis of the SEM image, the results indicate that when the glass sand with a larger particle size is mixed into the concrete as fine aggregate, due to the large amount of active SiO_2 in the glass sand, it reacts with OH^- , K^+ , and Na^+ in the concrete pore solution, resulting in the formation of loose ASR gel; then, the ASR gels are swelled by absorbing water, which increases the expansion rate of the specimen. When the particle size of ground glass is less than a certain value, the potential hydraulic activity and potential pozzolanic property of the glass powder are stimulated, and when used as auxiliary cementing material, the active ingredients in the glass powder such as SiO_2 are more involved in the pozzolanic reaction, consuming a large amount of $Ca(OH)_2$ and forming a large amount of dense CSH gels. On the one hand, because the OH^- in the solution is consumed in large quantities, the amount of OH^- participating in the ASR is reduced, thus reducing the amount of ASR gels and inhibiting ASR swelling; on the other hand, the generated CSH gels have a strong

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ability to fix Na⁺ [26], which reduces the Na⁺ concentration of pore solution and further reduces the ASR degree of active aggregate.

Therefore, combined with the results of expansion rate and SEM diagram analysis, it is inferred that the inhibition mechanism of glass powder on ASR expansion may be due to the potential pozzolanic property of ground glass powder. When glass powder is mixed into concrete as an auxiliary cementitious material, SiO₂ in glass powder is more involved in the pozzolanic reaction, consuming OH⁻, in solution and reducing ASR rate, thus reducing the formation of ASR gels. It plays a key role in inhibiting the expansion of ASR [7].

4. Conclusions

By studying the inhibition effect of waste glass powder on ASR expansion, comparing the expansion rate of concrete specimens with different amounts and particle sizes of glass powder at different ages, the effects of glass powder content and particle size on the inhibition of ASR expansion were analyzed. The ASR expansion effects of glass concrete and its mechanism were studied. The main results are as follows: (1) the main occurrence when usingglass sand is ASR, which produces fluffy ASR gel, leading to the increase inexpansion rate; the expansion rate of concrete increases with the increase of glass sand content, and the 14-day-old expansion rate of mortar bars mixed with glass sand is larger than the standard limit value; therefore, there is a considerable hidden safety danger when waste glass is directly used in concrete in the form of glass sand; (2) the glass powder produces dense CSH gel by pozzolanic reaction, and the consumption of a large amount of OH⁻, required by ASR, has a good inhibitory effect on the ASR expansion of concrete, and the inhibitory effect becomes more obvious with the increase in the fineness of the glass powder. The optimum content of glass powder in concrete is less than 20%, and the particle size of glass powder is less than 75 µm; (3) glasspowder and glass sand can be used in concrete as auxiliary cementitious material and aggregate, respectively, which is beneficial to the recycling of waste glass, but the content of glass sand should not be more than 20%.

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