Review



State of the Art on Waste Glass Powder as Supplementary Cementitious Material

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Abstract: As a new type of high-performance concrete material, glass powder (GP) concrete is beneficial to the environment and concrete performance. However, the specific application method still needs to be studied. This paper introduces the structure theory of glass and the mechanism by which glass powder affects concrete. Through experiments, the effects of glass powder particle size, content, alkali content, and other factors on alkali-silica reaction (ASR) expansion, mechanical properties, and durability of concrete are compared. It is proven that glass powder can be used as a gel material in concrete. The existing problems in the research are summarized, and the latest research perspectives and methods are suggested.

Keywords: waste glass powder, concrete, supplementary cementitious materials, alkali-silica reaction, mechanical properties, chloride penetration resistance

Abbreviations

GP	Glass Powder
WGP	Waste Glass Powder
WG	Waste Glass
C-S-H	Calcium Silicate Hydrate
ASR	Alkali Silicate Reaction
САН	Calcium Aluminate Hydrate
SEM-EDS	Scanning Electron Microscope and Energy-Dispersive X-ray Spectroscopy

1. Introduction

The rapid economic development sees a great increase in glass production and consequently, the waste glass production is also increasing (Figure 1). And as shown in Figure 2, the WG has high recover value. In 2020, the output of WG in China reached 22.12 million tons [1]. According to the relevant statistics of the United Nations, the proportion of WG in solid waste has reached 7% [2]. The recycling rate of WG in developed European countries is about 76% [3]. According to the Development Report of China's Renewable Resources Recycling Industry released in 2020 [4],

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China's WG recycling volume is about 8.6 million tons, and the recovery rate is only about 46.3%. The high melting point of glass and the high cost of collection, transportation, incineration, and difficult natural degradation through landfill, coupled with the difficulty of recycling and the relatively small economic benefits of glass recycling result in a low WG recycling rate in China. How to increase the use of WG and improve the use of glass efficiency is the breakthrough point in solving this problem.

Nowadays, recycling is a common practice because it preserves the earth's resources. Waste glass can be used as a substitute for supplementary cementitious materials in concrete. The optimal mix of recycled GP as an alternative to supplementary cementitious material and its immediate and long-term impact on the strength and durability of concrete are still being studied. However, it is expected to be a excellent candidate for reducing the cement portion in concrete.



Waste glass production and growth rate in China from 2014 to 2020

Figure 1. Waste glass production and growth rate in China from 2014 to 2020 [1]



Value and growth rate of waste glass recycling in China from 2014 to 2020

Figure 2. The value and growth rate of waste glass recycling in China from 2014 to 2020 [1]

In the construction industry, recycled WG is commonly used as grinding material, decorations, welding flux, or as additives. Concrete is the most used material in the construction production. How to effectively combine WG and concrete has become the bone of contention. According to the latest theoretical and practical data [5]-[7], the glass itself contains a large amount of SiO₂, Al₂O₃, Na₂O, CaO, and other chemical components, with potential hydraulic activity and pozzolanic activity, which can be used as raw materials to be added to concrete. This method is not only economically and technically possible but also results in higher-performance concrete. The main use of glass in cement concrete is divided into two categories, one is added to the concrete as an aggregate, and the other is used as a gel material. Due to the high Alkali content in the glass, whether WG is used as coarse aggregate or fine aggregate, the ASR phenomenon will occur [8], [9], which is detrimental to the mechanical properties of concrete. However, when the diameter of the glass grain is less than a certain value, that is, when it is added as GP, a Pozzolanic reaction will occur, and the calcium silicate hydrate gel (C-S-H) generated can not only enhance the structural strength but also the expansion of ASR in concrete [10]-[13]. Although it is very convenient to use WG as aggregate in concrete, this paper will not discuss it in view of its impact on the mechanical properties of concrete and its low usage efficiency. It will focus on the efficient application prospect of WG as a gel material.

This review article systematically organizes the research outcomes related to the behavior of concrete with glass powder such as mechanical properties, durability, and ASR expansion. The reaction mechanism was analyzed via a chemical reaction equation. The effects of glass powder content, particle size and alkali content, and other factors on the performance of concrete were studied based on its mechanical properties, ability of chloride penetration resistance, and acid resistance, etc.

2. Characteristics of glass powder

2.1 Classification and structure of glass

Composition (%)	SiO ₂	Al_2O_3	Fe ₂ O ₃	CaO	MgO	SO_3	Na ₂ O	K ₂ O	TiO ₂	Cr ₂ O ₃
Cement	20.8	4.6	2.8	65.4	1.3	2.2	0.31	0.44	-	-
Sand	88.54	1.21	0.76	5.33	0.42	-	0.33	0.31	0.05	-
Brown Glass	72.08	2.19	0.22	10.45	0.72	-	13.71	0.16	0.1	0.01
Green Glass	71.22	1.63	0.32	10.79	1.57	-	13.12	0.64	0.07	0.22
Clear Glass	72.14	1.56	0.06	10.93	1.48	-	13.04	0.62	0.05	-
Flat Glass	72.16	0.68	0.12	7.8	4.38	0.21	0.21	14.46	-	-
Hollow Glass	69.89	1.92	1.05	12.31	1.34	0.14	0.16	13.18	-	-
Windshield Glass	70.11	1.86	1.04	11.67	1.37	0.11	0.07	13.76	-	-
Windshield Glass	70.11	1.86	1.04	11.67	1.37	0.11	0.07	13.76	-	-

Table 1. Chemical composition of cement, sand, and different glasses [16]

The conventional glass production method is to melt a variety of inorganic minerals such as quartz sand, borax, boric acid, barite, barium carbonate, limestone, feldspar, soda ash, and other raw materials at 1550-1600 °C [14]. After adding a small amount of auxiliary raw materials, the glass is molded and cooled, and finally, the solidified transparent amorphous inorganic material is obtained. Glass can be categorized according to its different compositions, such as siliceous glass, alkali silicate glass, soda lime glass, borosilicate glass, lead glass, barium glass, and aluminum silicate glass, etc. The most common glass is soda lime glass [15]. The chemical composition of glass is shown in Table 1. It can

be seen from Table 1 that SiO_2 is the main component in glass, followed by a large amount of alkali, and X-ray detection shows that glass is a typical amorphous material [16].

Crystallography theory and irregular network theory are two famous glass structure theories in the current academia [17]. The crystallographic theory was proposed by Randall in 1930, who believed that the components of glass are made up of 90% microcrystalline and irregular network structures [18]. The latter, proposed by W. H. Zachariasen in 1932, argues that the theory of irregular network structures should be studied from the perspective of structural chemistry-chemical bonds. Zachariasen et al. proposed that glass and crystal similar to SiO₄ is the smallest unit of composition, a completely irregular arrangement connected to form a continuous three-dimensional network [19]. From the perspective of chemical bonds, Zahariansen believes that the oxygen ions in the glass connect the 'bridging oxygen' of the two network-forming cations as the network-forming oxide; the 'non-bridging oxygen' connecting the network is an intermediate oxide [19]. Hence, glass is mainly composed of these three oxides.

2.2 Factors of concrete affected by glass powder

The incorporation of GP mainly affects the mechanical properties, durability, and ASR expansion of concrete. Several studies have shown that GP can effectively inhibit ASR expansion as a gel material in concrete [20]. The inhibition effect is closely related to the fineness of GP. When the diameter of the GP is lower than a certain value and up to a certain limit, the expansion will not occur. Many studies have shown that the addition of GP to concrete can also improve its mechanical properties, and there is an optimal value due to the influence of particle diameter, incorporation amount, and age [21]. In addition, studies have shown that the incorporation of GP is also conducive to reducing the porosity of the microstructure and reducing the penetration of various solutions, thereby enhancing its durability [22]. As shown in Table 1, GP contains a large amount of silicon and calcium. Theoretically, once the GP attains certain fineness, it would achieve volcanic ash activity [23], [24]. The pozzolanic reaction is dependent on the particle size of GP and reaction temperature, that is, the smaller the particle size and the higher the temperature, the more intense the reaction [20].

2.3 Chemical excitation of glass powder in concrete

With the addition of only GP, the amount of cement clinker in the system is reduced, and due to the potential activity of GP, the hydration rate of cement in the early stage is correspondently reduced and resulting in a smaller amount of hydration products generated. The resulting system structure of the concrete is not dense, thus affecting its strength at an early stage. At this stage, the pozzolanic activity of GP is not strong. It mainly plays the role of physical filling. In construction practice, the addition of GP requires a longer curing process and this would affect the construction progress. In the later stage, SAR gradually plays a greater role. The active components in the GP and the Ca(OH) $_2$ in the hydration process undergo a secondary hydration reaction to form C-S-H and calcium aluminate hydrate (CAH). As more and more GPs participate in the hydration reaction, it improves the pore structure of the cementitious system, compactness, and performance of the concrete. The active SiO₂, active Al₂O₃, and other components are hydrated with the clinker in cement to form Ca(OH)₂, which generates C-S-H, CAH, or hydrated calcium aluminate sulfate. The so-called pozzolanic Portland cement reaction and the reaction chemical equation are as follows:

$$SiO_2 + Ca^{2+} + 2OH^- \rightarrow n_1CaO \cdot SiO \cdot 2n_2H_2O(s)$$
 (C-S-H)

At present, the common chemical excitation methods are alkali excitation, salt excitation, and salt-alkali composite excitation. The most common is the addition of alkali activator. Under the action of OH⁻, the Si-O bond and Al-O bond on the surface of WGP particles are broken, which reduces the degree of polymerization of the Si-O-Al network and forms free unsaturated active bonds on the surface, causing it to hydrate easily with Ca(OH)₂ to form C-S-H and CAH gels. However, excessive alkali will also cause ASR expansion, so it is still necessary to explore the optimal ratio. The commonly used alkali activators include Mn $\{-(SiO_2)_rAIO_2\}n \cdot wH_2O$ and common salts, such as Na₂SO₄, etc.

3. Application of glass powder in concrete

GP has three major effects on concrete mechanical properties, impermeability, and ASR expansion. At present, the research direction of GP in academic circles focuses on investigating the relationship between calcium-silicon ratio, expansion rate, product, and structure in volcanic ash reactions by thermogravimetric analysis, scanning electron microscope, and energy spectrum analysis (SEM-EDS) [25]-[27]. In addition, researchers [28] also measured the concentration of various ions in the pore solution of various phases by inductively coupled plasma to infer the hydration process of the system and assisted with scanning electron microscopy and SEM-EDS to explore the effect and mechanism of WGP as an auxiliary cementitious material on mortar performance [28].

3.1 Alkali-silica reaction

The alkali-silica reaction mechanism is $Na^+(K^+) + SiO_2 + OH^- \rightarrow Na(K)$ -Si-H gel. The volume of alkali silicate gel after water absorption is much larger than that of before the reaction, and the maximum volume can be increased by more than 3 times. A large number of gels accumulate and expand in the interface area of the concrete aggregate, resulting in uneven expansion and cracking of concrete at the interface. This is a common phenomenon in engineering. The local expansion during cement hydration and solidification leads to concrete rupture that is difficult to repair. The GP is added to the concrete as active silica, and the dispersed active silica can be uniformly reacted during the hydration curing process to avoid local expansion [29].

A large number of experimental studies have shown that the inhibitory effect of GP on ASR is related to the particle size, dosage, and alkali content of GP.

3.1.1 Particle diameter



Figure 3. ASR expansion of the mortars containing WGPs with different particle sizes [34]

Huang et al. [30] and Hendi et al. [31] mixed glass sand with a particle size of 5 mm and micron GP into concrete in a certain proportion, and found that the former promoted the expansion of ASR to some extent. On the contrary, the volcanic ash reaction of micron GP can inhibit the occurrence of ASR. Wang et al. [32] believe that when the particle size of GP is less than 300 μ m, the expansion is at a very low level. Wang [33] also proved by measuring the length of mortar bar mixed with GP with different grinding times that the longer the grinding time of glass, the smaller the particle size, the greater the shrinkage value of the corresponding mortar bar, that is, there is a significant inhibitory effect. Li et al. [28] and Liu et al. [34] also proved through experiments that the finer the particle size of WGP, the smaller the expansion rate of mortar. The expansion rates of 0-0.075 mm and 0.075-0.15 mm were reduced by 20.2% and 8.4%, respectively. When the particle size of WGP is less than 0.15 mm, the effect of ASR expansion can be significantly reduced, as shown in Figure 3. But some researchers [30], [33] also pointed out that the GP is not the finer the better. There is an ASR expansion of the GP particle size limit. Wang et al. [35] also pointed out that the particle size of WG is mostly $5 \sim 20 \,\mu\text{m}$, and there is still a lack of research on the influence of ultrafine GP below 5 μm or even submicron on the reaction characteristics of cement paste. Therefore, in practice, the study of GP to inhibit ASR expansion should focus on the optimal particle size range, and academically, the effects of particles smaller than 5 μm can continue to be explored.

3.1.2 Dosage of glass powder

As mentioned above, when GP is added to the concrete as an auxiliary gel material, the first volcanic ash reaction reduces the content of Ca(OH)₂ in the reactants, thereby reducing the occurrence of the ASR. Multon et al. [36] think that it is due to the colloid generated by the reaction, which fills the voids of the concrete aggregates, thereby inhibiting the occurrence of expansion. With the increase in the tempered GP content, the more colloid filled the gaps in the concrete aggregates, the smaller the expansion value of the mortar bar. Hendi et al. [31] and Wang et al. [37] showed that when the WG was fine enough, the ASR reaction of concrete decreased by 5%, 14%, 23%, and 52% respectively when the substitution rate was 5%, 13%, 20%, and 30% respectively, as shown in Figure 4. Other studies [20], [29], [33], [38] have shown that, with GP instead of cement or as a gel material added to concrete, ASR expansion decreases with the increase of GP content and even could shrink. In addition, Shi et al. [39] also found that salt or alkali can be added as an activator to improve the efficacy of GP. Experiments [39] show that the best replacement rate of inactive WGP is 10%, and the expansion rate of mortar is better than the reference group. When the replacement rate of WGP is 30%, the expansion rate of mortar is higher than 0.14%, which has a potential ASR expansion risk. Therefore, the amount of GP, but there is also a certain limit. In practice, the amount of WGP should be controlled.



Figure 4. The expansion rate of ASR of mortars with WGP substituting 0, 10%, 20%, and 30% of Portland cement [38]

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3.1.3 Alkali content

The alkali contained in WG is the main cause of the alkali-silica reaction. Researchers [40] calculated the total alkali content in WG by the equivalent amount of sodium oxide. The formula is expressed as alkali content (%) = Na_2O (%) + 0.658K₂O (%). From the chemical formula of soda lime glass (containing 13% Na_2O), it can be calculated that the alkali content of GP accounts for a large proportion. But ASR does not occur in solid alkali, so the alkali in GP not all participates in the reaction. It becomes an active component in a dissolved state and GP can only release a small amount of alkali into the solution [41]. Researchers [42] used GP to replace part of high alkali cement to configure concrete and found that the expansion value of concrete was less than 0.02%, that is to say, GP would not cause harmful expansion in the absence of active aggregate. In addition, as shown in Figure 5, some researchers [39] found that the occurrence of ASR can be better inhibited by adding salt- to promote the activity of GP, and there is a certain threshold for the optimal modulus and concentration of activator, which can be the research direction of future experiments.



Figure 5. ASR expansion of the AAS mortars containing different alkali dosages [39]

3.2 Mechanical properties

The application of GP as a gel material in concrete can improve its mechanical properties of concrete to a certain extent. There have been a large number of experiments on the performance of GP concrete at home and abroad. The independent variables of the experiment are mainly cement and GP. The cement test direction mainly includes cement paste, cement mortar, and concrete test block. The research directions of independent variable GP include GP types (white glass, green glass, tempered glass, etc.), particle size, dosage, alkali content, and test block curing time. The dependent variables mainly include the compressive strength, flexural strength, and workability of concrete.

Researchers [43], [44] replaced cement with 5%, 10%, 15%, 20%, and 25% WGP to test its effect on concrete strength. The results are close, and one set of data is shown in Table 2. The test results show that the compressive strength of concrete with GP at 28 d is greater than that of ordinary concrete, and the strength of GP concrete reaches the maximum when the substitution rate is 15%. In addition, the mechanical properties of concrete are also related to the GP diameter. The smaller the particle size of GP is, the higher the pozzolanic activity will be. Micron-sized GP can significantly enhance the mechanical properties of concrete [45]-[49]. From Figure 6 [50] and Figure 7 [51], we can draw the conclusion that the application of GP as a gel material in concrete can improve the mechanical properties of

Mixture	Sample Number	Tensile Strength (MPa)	Average Tensile Strength (MPa)	Standard Deviation (MPa)	Variability Index (%)
CONTR	1.1–1.9	5.81-10.19	8.3	0.78	9.36
CG10	2.1-2.9	5.21-7.91	6.9	0.69	10.02
CG30	3.1-3.9	6.08-8.23	7.43	0.39	5.19
CG50	4.1-4.9	3.6-5.67	4.55	0.63	13.79
CG100	5.1-5.9	2.51-5.17	3.84	0.73	19.02

Table 2. Results of compressive strength tests for concretes [44]





Figure 6. The compressive strength of waste glass concrete [50]

The strength of the test blocks increases with the increase in curing time, which is consistent with the conclusions of [52], [53]. This is because GP promotes the early hydration reaction and improves the later hydration process as a whole.

Chen at al. [21] used electron microscope scanning to analyze the influence of GP on cement mortar at the micro level, as shown in Figure 8 and Figure 9. The observation results show that the cement mortar with GP has a denser structure and corresponding stronger mechanical properties. The generated C-S-H gel is shown to fill the structure, which is the same as the experimental results of Sobolev et al. [54]. Shayan et al. [55] and others' experiments also proved that the pozzolanic reaction of GP will decrease Ca (OH)₂ content in cement. Combined with Taha et al [56], the expansion rate mechanism of the alkali-silica reaction of GP applied to cement is explained. It is proven that glass

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powder can reduce the ASR reaction and improve the structural strength of concrete by reducing the alkali content in concrete.



Concrete mix ID

Figure 7. The compressive strength of concrete mixes [51]

In addition, some researchers [57]-[61] pointed out that GP mixed with other admixtures can further improve the mechanical properties of concrete. This is consistent with the conclusion with Song et al. [62]. By adding a salt-alkali activator, mechanical properties of concrete were enhanced. A large number of experiments have proved that GP can indeed improve the mechanical properties of concrete. However, the optimal substitution ratio needs to be studied in combination with specific glass types, GP size, dosage, alkali content, etc. Finding more efficient activators is also the subject of future research in related fields.



Figure 8. SEM diagram of the benchmark group [21]. (a): Microstructure of block with curing time of 3 days; (b): Microstructure of block with curing time of 7 days; (c): Microstructure of block with curing time of 28 days

3.3 Durability

The durability of concrete is mainly reflected in the resistance to chloride ion penetration and acid resistance. Chloride ions could erode the steel bars in reinforced concrete, and react with them. Researchers [22], [63] simulated

the marine environment to test the influence of seawater erosion on concrete. The results show that a large number of chloride ions can penetrate into the concrete within 28 days, resulting in permanent defects in concrete. Through the prediction of concrete life, its remaining service life is only 12 years. As shown in Figure 10, the life curves of concrete with different GP and silica fume contents show the same change rule. Researchers [54] found that improving the resistance to chloride ion penetration of concrete is mainly through improving its pore structure, reducing the emergence of large pore bubbles, hence making the concrete denser and more durable.

Through the composite solution of four kinds of salts, the optimum content of GP in the full immersion test of GP/ silica fume was studied. The experimental results show that when the content of GP is 10% and the content of silica fume is 6%, the durability of concrete is the optimum proposal. Jain et al. [64] mixed GP and granite powder to replace cement and found that when the replacement rate of the two was 15% and 30%, respectively, the resistance of concrete to chloride ion penetration was greatly improved. Wu et al. concluded that a denser structure can reduce chloride ion permeability. Reference [65] replaced 10% and 20% cement with 5 μ m and 12 μ m GP respectively, and found that the higher the substitution rate and the smaller the particle size, the lower the chloride diffusion coefficient of concrete. Academia often applied electric field calculation of the ability of chloride penetration resistance. The formula [66] is as follows:

$$\mathbf{DRCM} = \frac{0.0239 \times (273 + T)L}{(U - 2)t} \times \left(Xd - 0.0238 \sqrt{\frac{(273 + T)LXd}{U - 2}} \right)$$

In the formula: **DRCM** is the unsteady chloride ion migration coefficient of concrete; U is the absolute value of the applied voltage; T is the average value of initial temperature and final temperature of the anodic solution; L is the thickness of the test block; Xd is the calculated average chloride penetration depth; and t is the experimental time.



Figure 9. SEM diagram of the group with 40% glass powder [21]. (a): Microstructure of block with curing time of 3 days; (b): Microstructure of block with curing time of 7 days; (c): Microstructure of block with curing time of 28 days

In terms of acid resistance, Mostofinejad et al. [67] prepared concrete by substituting cement partly with GP for sulfate exposure experiments. The results show that the acid resistance of concrete is 53% higher than that of ordinary concrete. At the same time, Harbec et al. [68] compared the acid resistance of WGP mortar and silica fume mortar with the same substitution rate, and found that the early sulfate resistance of WGP mortar was weaker than that of silica fume mortar because the pozzolanic reaction of WGP was slower than that of silica fume. Djomo et al. [69] believed that the secondary hydration products are produced by the incorporation of WGP. GP reduced the porosity of concrete, thereby improving the acid resistance of concrete. Jain et al. [64] also showed that the sulfuric acid corrosion resistance of concrete.



Figure 10. Erosion life curve of different concrete composite salt solutions [22]

4. Application of glass powder in concrete

Many experiments have proven that WGP has a wide application prospect as a concrete gel material. However, ordinary grinding methods cannot effectively break the strong chemical bonds in WG, the activity of WG even in the powder state remains relatively low [40]. Although many scholars have improved the activity of GP by adding activators or other admixtures, exploring the most effective direction to improve the activity of GP remains the focus of future research.

With the rapid development of high-performance computing devices, it is feasible to simulate amorphous C-S-H gel at a molecular scale using molecular dynamics method. In recent years, domestic and foreign scholars have studied the toughening effect of graphene oxide, carbon nanotubes, and other nanomaterials on C-S-H gel at a molecular level. The demand for the study of the unsaturated transport of ions and water molecules in C-S-H nanopores is increasing year by year. However, the relatively complex structure of C-S-H and the diverse interface configurations of graphene oxide- and C-S-H limit the experimental study of micro/nano-scale mechanisms, especially at the molecular level. The question to be asked is if it is possible to use the molecular dynamics method and configuration design theory to construct an anisotropic hydrated calcium silicate and GP/C-S-H interface optimization model. Such approach aims to adjust the model structure and interface interaction to improve the overall performance of composite materials.

Although scholars have done a lot of research on the microstructure, composition, and properties of C-S-H gels, it is impossible to comprehensively analyze the properties and microstructure of C-S-H gels due to their small particle size distribution, uncertain chemical composition, poor crystallinity, and amorphous state at room temperature. Experimental techniques, such as atomic force microscopy, X-ray diffraction, scanning electron microscopy, solid-state nuclear magnetic resonance, neutron diffraction, and so on, can accurately test C-S-H gels on the scale of 10⁻¹-10⁻⁹ m. However, the theoretical calculation method can more intuitively characterize the microstructure of cement-based materials at the scale of 1-100 Å, simulate the movement of a large number of molecules and ions, obtain the kinetic information in the diffusion process, and effectively assist the experimental technology.

In addition, the practical application experiment of GP concrete is still relatively simple. In the future, the advantages of frost resistance, high-temperature resistance, and carbon neutralization of GP concrete can be further studied.

5. Conclusions

In this research work, taking glass powder as supplementary cementitious material and comparing its impact on the performance of concrete and problems arising from the current research, the following conclusions can be drawn:

(1) GP has unique hydration characteristics in the Portland cement environment, and its application in concrete as a new type of auxiliary cementitious material has good application prospects.

(2) The GP used in concrete can effectively inhibit the occurrence of ASR expansion and is related to the diameter of the GP, content, and alkali content.

(3) The application of GP as a gel material in concrete can improve the mechanical properties of concrete to a certain extent, which is related to the particle size, content, alkali content, and curing time of the test block.

(4) GP, as a gel material used in concrete, can effectively improve the durability of concrete.

(5) When GP is mixed with other admixtures, it can improve the mechanical properties of concrete better than using GP alone. It is still worth further study to explore more effective GP admixtures.

(6) With computers, the molecular dynamics method and configuration design theory can be used to study the action process of materials at the microscopic level.

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Conflict of interest

The authors declare no competing financial interest.

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