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Retraction

Retracted: Application of Foam Glass-Ceramic Composite Thermal Insulation Material in Traditional Buildings

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This article has been retracted by Hindawi following an investigation undertaken by the publisher [1]. This investigation has uncovered evidence of one or more of the following indicators of systematic manipulation of the publication process:

(1) Discrepancies in scope
(2) Discrepancies in the description of the research reported
(3) Discrepancies between the availability of data and the research described
(4) Inappropriate citations
(5) Incoherent, meaningless and/or irrelevant content included in the article
(6) Peer-review manipulation

The presence of these indicators undermines our confidence in the integrity of the article's content and we cannot, therefore, vouch for its reliability. Please note that this notice is intended solely to alert readers that the content of this article is unreliable. We have not investigated whether authors were aware of or involved in the systematic manipulation of the publication process.

Wiley and Hindawi regrets that the usual quality checks did not identify these issues before publication and have since put additional measures in place to safeguard research integrity.

We wish to credit our own Research Integrity and Research Publishing teams and anonymous and named external researchers and research integrity experts for contributing to this investigation.

The corresponding author, as the representative of all authors, has been given the opportunity to register their agreement or disagreement to this retraction. We have kept a record of any response received.

References

[1] Q. Yu, "Application of Foam Glass-Ceramic Composite Thermal Insulation Material in Traditional Buildings," *Journal of Chemistry*, vol. 2022, Article ID 9662805, 9 pages, 2022.

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Research Article

Application of Foam Glass-Ceramic Composite Thermal Insulation Material in Traditional Buildings

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In order to solve the application of thermal insulation materials in traditional buildings, a foam glass-ceramic composite thermal insulation material was proposed for the application of traditional buildings. First, the preparation of the hydrated glass matrix was completed by using the colloidal chemical method with sodium water glass, boric acid, calcium bentonite, and fly ash as the main raw materials. Second, using the introduction of crystalline mineral powder fly ash and secondary heat treatment, a low-temperature foam glass-ceramic composite material with lightweight, high strength, good water resistance, and low cost is prepared. Finally, the process parameters such as dehydroxylation heat treatment temperature and holding time involved in the preparation process were explored and optimized. It is proved that the external content of fly ash is 20 wt.% and above, and the external content of boric acid is 1-2 wt.%. The mixed sol has a bulk density of 192–256 kg/m^2, a compressive strength of 0.44–0.63 MPa, a weight loss rate of 5.1–7.8 wt%, and a softening coefficient of 0.85–0.95, and thermal conductivity is 0.056–0.064 W/(m·K).

1. Introduction

In recent years, with the continuous introduction of new foam glass products by manufacturers of foam glass-ceramic composite thermal insulation materials in countries such as Germany and Japan, the market demand for foam glass has suddenly increased. In the early 1990s, the market demand for foam glass in China was only 6,000 m^3. In the mid-1990s, the market demand was about 8,000 m^3. By the end of the 1990s, the market demand for foam glass had risen to 20,000 m^3. Experts predicted that the annual demand for foam glass would reach 1.0×10^5 m^3 by 2010, and the demand for foam glass in the domestic market would show a rising trend of both production and sales. Foam glass manufacturers should use this favorable opportunity to vigorously develop, promote, and export high-quality foam glass products, so that the development and utilization of domestic wall building materials will produce qualitative changes, and high-quality, multivariety foam glass will enter the international market [1]. At present, there are small factories in China which use waste glass to produce foam glass products. The annual production capacity of these small factories is only a few thousand cubic meters. The technical equipment is very backward, the product quality is unstable, the scrap rate is high, and the cost remains high, which hinders the promotion and use of the product. Despite this, the product is still in short supply.

Although there are many varieties of existing thermal insulation materials, they are very limited as building thermal insulation materials. Judging from the actual application of the existing building thermal insulation materials, there are certain defects. From the perspective of the performance of these materials, there are problems such as high water absorption, low strength, no fire resistance, and inconsistent expansion coefficients with concrete or brick wall materials. Judging from the actual situation after the thermal insulation construction, the cracks and hollowing of the wall surface are serious. Because most of the materials are organic, they are easy to age and have a short service life. These not only affect the overall appearance of the wall but also have extremely difficult maintenance and inconvenient issues, as shown in Figure 1. On the basis of the current research, a foamed glass-ceramic composite thermal insulation material is proposed for use in traditional buildings. After a lot of exploration in the previous experiments, it was found that the water resistance of the low-temperature foamed glass-ceramic composite material was significantly improved when the high-temperature dehydroxylation heat treatment was completed at 525°C and higher.

2. Literature Review

With the rapid development of science and technology and the increasing improvement of people's living standards, foam glass-ceramic composite thermal insulation materials are widely used in housing construction and people's daily life and have become an indispensable material for scientific experiments and cutting-edge technologies. Therefore, a lot of waste glass is inevitably produced, and various waste glass occupies a certain proportion in glass factories and municipal waste. According to statistics, in some developed countries in Europe and in the United States, waste glass accounts for 4% to 8% of the total urban waste. About 3.2 million tons of waste glass are produced every year, accounting for 2% of the total municipal solid waste [2]. In the past, most of the waste glass was recycled by some Xiaoping glass factories, but the production process of Xiaoping glass is backward, the equipment is simple, the environmental pollution is serious, and the energy consumption is high. The flat glass produced has low light transmittance, and there are a lot of defects such as waves, bubbles, pitting, and scratches; and the glass is brittle and easily broken. In order to change this situation, these small flat glass production lines have been phased out within a time limit. Therefore, how to rationally reuse waste glass has important practical significance. The use of waste glass to produce foam glass is an effective way to solve this problem. Foam glass is mainly made of waste glass and various glass-containing substances. After crushing and grinding, adding foaming agents, modifiers, and other materials, and uniformly mixing to form batches, it is then placed in a specific mold to undergo melting, foaming, and annealing to form a porous glass material filled with uniform pores.

Sayan et al. used waste glass as the main raw material, borax as a flux, and carbon powder as a foaming agent and then added certain additives to prepare microcrystalline foam glass [3]. Melnikov et al. applied mathematical analysis methods to establish a relatively accurate mathematical relationship between the pore diameter and temperature of foam glass and improved its preparation research theory [4]. Shang prepared a foam glass material for military use to absorb radar waves, and the research on foam glass turned to the development of various new functions [5]. Lu and Shi used silica sand, soda ash, feldspar, and so forth as the basic raw materials, plus a small amount of foaming agent and crystal nucleating agent, sintered according to a certain system, and slowly cooled to prepare microcrystalline foam glass [6]. Liu et al. used hexachlorocyclotriphosphorus cyanide graft-modified expandable graphite as a flame retardant and added it to isocyanate and polyether, mixed and foamed to obtain a flame-retardant rigid polyurethane foam, which improved the LOI and prolonged the ignition time; the performance is improved compared with pure rigid polyurethane foam [7]. Peng adopted the organic foam impregnation method and foamed gel process, externally doped with cerium phosphate and yttrium oxide for toughening to prepare siC-based foam ceramics, and optimized the slurry preparation and sintering system [8]. Chen et al. used fly ash, waste glass powder, and ceramic powder as the main raw materials, sulfate as foaming agent, and boric acid as foam stabilizer. A product with a bulk density of 645 kg/m^3 and a compressive strength of 8.01 MPa was produced. Although the product has high strength, its thermal insulation performance is poor [9]. Buriachok et al. used coal gangue and shale as the main raw materials and sintered them at 1200°C by powder sintering method to obtain a foam product with a thermal conductivity of 0.14 W/(m·K) [10]. Nguyen et al. used waste slope glass and water as the main raw materials to prepare foam slope glass by hydrothermal hot pressing-calcination method and finally obtained a foam slope glass product with a bulk density of 250 kg/m^3 and a compressive strength of 1.5–1.6 MPa [11]. Lu used the waste of foamed glass as the main raw material, mixed with part of fly ash, and used hydrogen peroxide as the foaming agent. The foam glass products with thermal conductivity of 0.043–0.061 w/(nvk) were obtained, and the production cycle was as long as 7 days [12].

On the basis of the current research, a foamed glass-ceramic composite thermal insulation material is proposed for use in traditional buildings. After a lot of exploration in the previous experiments, it was found that the water resistance of the low-temperature foamed glass-ceramic composite material was significantly improved when the high-temperature dehydroxylation heat treatment was completed at 525°C and higher. The series of mixed sols prepared by the optimized formula will complete the low-temperature foaming heat treatment at 400°C and then they will be directly placed at a dehydroxylation heat treatment temperature of 525°C for 30 min. It was found that the water resistance of the material was significantly improved, the weight loss rate was reduced to less than 5 wt.%, and the softening coefficient reached more than 0.9.

3. Application of Foam Glass-Ceramic Composite Thermal Insulation Material in Traditional Buildings

3.1. Foam Glass-Ceramic Composite Thermal Insulation Material

3.1.1. Introduction. Foam glass is made of glass as the base material, which is foamed and fired at high temperature by ball milling. It is divided into closed-cell foam glass and open-cell foam glass. The walls are insulated with closed-cell foam glass. Closed-cell foam glass is a lightweight, rigid material composed of millions of completely closed glass bubbles. Each airtight bubble can play a thermal insulation effect and is an inorganic environmental protection advanced thermal insulation material. Foam glass has excellent comprehensive properties, as well as strong weather

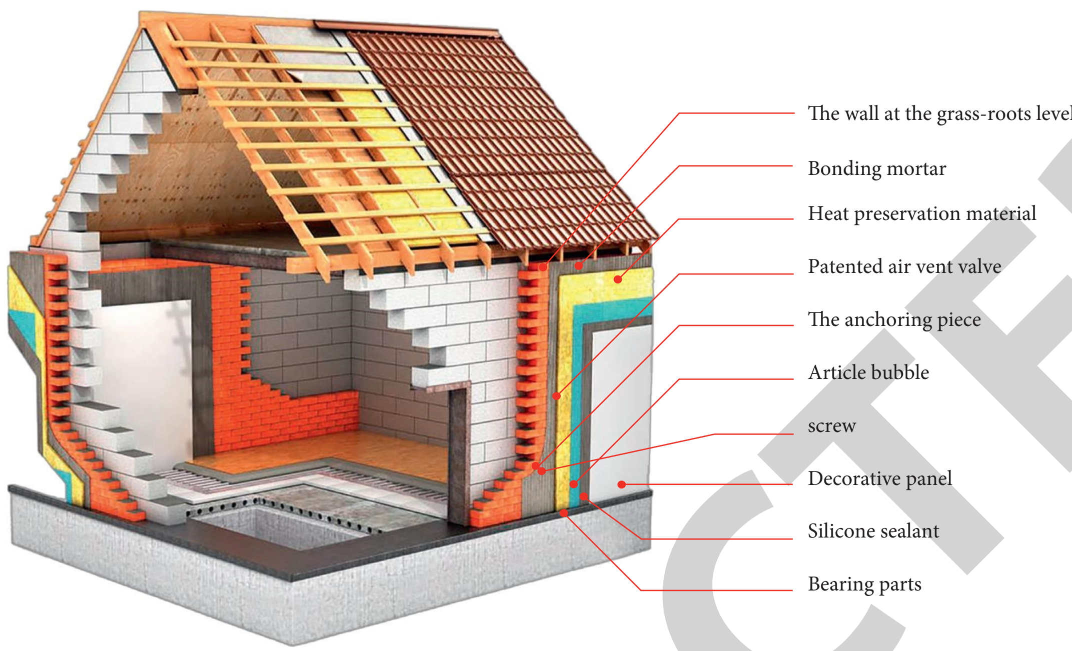

Figure 1: Composite thermal insulation material.

resistance and durability. It is an inorganic thermal insulation material. Its service life is synchronized with the building. The coefficient of linear expansion is basically the same as that of wall materials such as concrete. The surface is porous and the cement mortar is well bonded, and the construction is convenient. It has the characteristics of high strength, low thermal conductivity, and no water absorption. It has strong resistance to ultraviolet rays and wind and rain and has a permanent thermal insulation effect, which avoids the damage of the building structure due to changes in high and low temperatures [13].

3.1.2. Features. Foam glass thermal insulation material is an inorganic material and is firmly combined with other inorganic materials. It has good weather resistance, lightweight, high mechanical strength, good physical stability, and convenient transportation. It can be cut with common woodworking tools. It has good combination with ordinary cement mortar and is easy to paste on the wall. The construction of the material is simple and fast, and there is no need to set up a large area of reinforced mesh cloth and waterproof elastic putty, and the construction method is not limited by seasons. It can be pasted and installed by conventional methods. It is not only safe and reliable but also durable when used in harsh environments of low temperature and cryogenic temperature, underground engineering, flammation and explosion, humidity, and chemical erosion. It is known as "permanent thermal insulation material without replacement." Therefore, the foam glass wall insulation system is widely used in thermal insulation projects in civil and commercial housing construction, petroleum, chemical industry, underground engineering, shipbuilding, and defense and military industries.

3.1.3. Process Principle and Structure of Foam Glass Thermal Insulation Material. Foam glass has good dimensional stability. In actual use, there will be no cracks or deformation damage due to expansion and contraction. As long as a reasonable and matching protective layer and finishing layer are selected, the foam glass insulation system can have the same life as the building. When the foam glass is bonded to the base wall, the point-bonding method is used, and the coating area is required to be 40% to 70% of the area of the board. This not only effectively ensures the bonding strength of the foam glass plate and the wall but also forms a certain cavity, which is conducive to removing moisture. In the entire structural design of the foam glass exterior wall thermal insulation system, the compatibility of the materials in the system and the matching of the elastic modulus change indexes of the adjacent structural layers are fully considered. The special plastering mortar ensures a certain flexibility in order to release the deformation stress. Since the foam glass is a hard inorganic material, the mesh cloth is canceled in the ordinary structure, but the mesh cloth is added to disperse the deformation stress and prevent cracking when strengthening the structure or special parts such as door and window openings [14]. When making the paint finishing layer, the flexibility and deformation properties of the plastering mortar layer, putty layer, and paint layer are required to be gradually increased to prevent cracks in the finishing layer.

3.1.4. Comparison with Other Thermal Insulation Materials. At present, the applied building insulation materials mainly include rock wool boards, expanded perlite products, expanded vermiculite products, and foamed plastics. Although these thermal insulation materials have good thermal performance, they also have some disadvantages. For example, rock wool has a high water absorption rate, and its thermal conductivity is small when it does not absorb water. Once it absorbs water, its thermal conductivity will increase sharply. Moreover, due to the internal water absorption of rock wool, it is not easy to evaporate for a long time, which has a negative effect on the thermal insulation and heat insulation of buildings. The water absorption rate of expanded perlite products and expanded vermiculite products is also very large. There are many varieties of foam plastic. Foamed plastics for building energy saving mainly include polystyrene board, polyurethane board, and polyethylene board. Generally, the thermal conductivity of foamed plastics is lower than that of foamed glass, but the water absorption rate is relatively high. The water absorption rate of polystyrene board is more than 6%, and the water absorption rate of polyurethane board is more than 4%. The expansion coefficient of foam is much larger than that of cement or steel, and its dimensional stability is relatively poor. Compared with foam glass, foam plastic has poor fire resistance and cannot be used in parts with high fire protection requirements (such as external wall insulation). As an organic material, foam plastic also has problems of aging and failure. Foam glass is used for roof insulation and can also play a second waterproof role [15]. Building thermal insulation materials generally requires low water absorption, high strength, low expansion coefficient, and low thermal conductivity. With its excellent performance, foam glass is not only suitable for thermal insulation of building exterior walls and basements but also more suitable for roof thermal insulation. The experience of using foam glass as a building insulation material in the United States has proved that foam glass is durable and has excellent performance and quality.

3.2. Application of Foam Glass in Building Energy Saving

3.2.1. Building Roof Insulation. There are three main types of foam glass used for building roof thermal insulation structure: upright flat roof, inverted flat roof, and sloping roof. Inverted roofs can be used for greening, planting flowers, or as roof sports fields and other roofing. Foam glass is easy to combine with other inorganic materials and easy to construct and has better waterproof, fireproof, and thermal insulation effects. The actual engineering applications include the office building of the State Planning Commission, the residential building of the Central National Song and Dance Troupe, and the No. 14 residential building in the second district of Fangguyuan, Beijing.

3.2.2. Building Exterior Insulation. As a wall thermal insulation material, foam glass can effectively reduce the thickness of the wall, reduce the quality of the building structure, and expand the usable area. The use of foam glass as external thermal insulation technology has been adopted in Changchun and other regions.

3.3. Problems Existing in Research and Production of Foam Glass. Foam glass has not been widely used for building energy efficiency, mainly because of cost. In the early days, it was mainly used in the petrochemical industry and the price reached 2000–3000 yuan/m^3. For building insulation, if the price exceeds 1,500 yuan/m^3, it is difficult to be competitive with EPS sheets and polystyrene granular mortar. The reasons for the high production cost of foam glass are mainly as follows.

3.3.1. Production Scale. In the development process of the foam glass industry, due to the limited strength of the foam glass manufacturers and the lack of effective industry planning and guidance, the current production scale of foam glass is small and has not formed an industrial scale. For foam glass, the cost of fuel (energy consumption) is an important component, and the larger the production capacity of a single kiln, the lower the unit energy consumption [16]. If the annual kiln production capacity is increased from 300 m^3 to 10,000 m^3, the cost of blanks will correspondingly drop from 1,500 yuan/m^3 to about 400 yuan/m^3. However, the annual production capacity of most kilns is between 3000 and 5000 m^3, and less than 20 can maintain normal production.

3.3.2. Production Process. The current production of foam glass in China is the same as that in foreign countries. The glass, foaming agent, and additives are mixed together, ground into powder, and then heated and foamed in a kiln. According to the different preheating-foaming-stabilizing-annealing cooling firing processes, it can be divided into "two-step" process and "one-step firing" process. The characteristic of the two-step firing method is in the later stage of foaming and shaping. After the heat-resistant steel mold is removed outside the kiln, it is concentrated in the annealing kiln for annealing. The advantage of this process is that it can observe and judge the foaming quality in time, adjust the foaming temperature and time system in time, and separate the foaming kiln from the annealing kiln. The adjustable range of the front and rear process tables is large, and it can flexibly produce foam glass of different densities and varieties. At present, domestic manufacturers all adopt the "two-step method" production process. Whether it is the "one-step method" or the "two-step method" to produce foam glass, the powder is loaded into the mold and then sent to the foaming kiln. The mold and the powder are heated together and then cooled, which consumes a lot of energy [17]. In particular, the intermittent production unit consumes more energy. For example, the intermittent production of thermal insulation foam glass with an electric furnace accounts for more than 70% of the cost. At the same time, the foaming temperature of thermal insulation foam glass is above 800°C, and the mold is made of alloy steel, which is expensive and has a large loss after repeated high-temperature heating. Some enterprises use cast iron as molds. Although the price is low, it is oxidized, peeled off, and deformed after heating, and the loss is also serious. The energy consumption of the mold and the consumption of the mold itself are another important reason for the high cost of foam glass [18].

4. Research and Development of Low-Temperature Foam Glass-Ceramic Composite Building Insulation Material

4.1. Material Preparation Process. The preparation process of the low-temperature foam glass-ceramic composite insulation material is as follows: (1) preparation of multihydrated glass matrix, where a certain amount of glass matrix modifier (a kind of boric acid and calcium-based bentonite) is added to liquid sodium silicate by wet chemical method or shearing at a high speed for 10 min at a rotational speed of 4000 rpm/min to obtain Na_2O-B_2O_3-Al_2O_3-H_2O hydrated glass sol; (2) physically blending into crystalline phase mineral powder, where, introducing crystalline phase mineral powder fly ash into the Na_2O-B_2O_3-Al_2O_3-H_2O system hydrated glass sol, physical blending was carried out with a high-speed dispersing sand mill at 4000 rpm/min for 30 minutes, and a gray liquid mixed sol was obtained without gelling, and the viscosity was higher than that of liquid silicic acid; (3) low-temperature foaming heat treatment, where the mixed liquid sol was poured directly into a 100 mm × 100 mm × 50 mm alumina ceramic crucible mold, and it was moved to a program-controlled box-type resistance furnace; the heating rate is warmed up from room temperature to 400°C of insulation for 30 min and obtains the low-temperature foaming heat treatment sample where the residual hydroxyl content is about 5%; (4) high-temperature dehydroxylation heat treatment, where 400°C of low-temperature foaming heat treatments, demoulding, and cutting into several 40 mm × 40 mm × 30 mm cuboid test blocks is placed in an alumina porcelain boat and directly moved into an electric heating constant temperature blast drying oven that has been heated to 500°C for 60 min–240 min. After cooling in the furnace, the low-temperature foam glass-ceramic composite thermal insulation material was obtained [19].

4.2. Composition, Properties, Structure, and Formation Mechanism of Low-Temperature Foamed Glass Ceramics

4.2.1. Thermogravimetric Analysis. Sodium water glass, boric acid, and fly ash (particle size < 35 um) with a modulus of 2.33 and a solid content of 47.22 wt% are used as raw materials. The mixed sol was prepared according to the formula, and the thermogravimetric test was carried out after drying for 12 h to explore the weight loss during the heat treatment of the dried blank [20].

Boric acid was introduced into sodium water glass through high-speed shearing (4000 rpm/min for 30 min) to obtain Na_2O-B_2O_3-Al_2O_3-H_2O hydrated glass sol without gelation. Then, the fly ash is added to the sol and the physical blending is completed through high-speed dispersion to obtain a mixed sol. The mixed sol was dried in a blast drying oven at 100°C for 12 h to obtain a green body with a certain moisture content. At this time, the mixed sol mainly lost a part of free water. The thermogravimetric analysis was carried out, and the results obtained are shown in Figure 2.

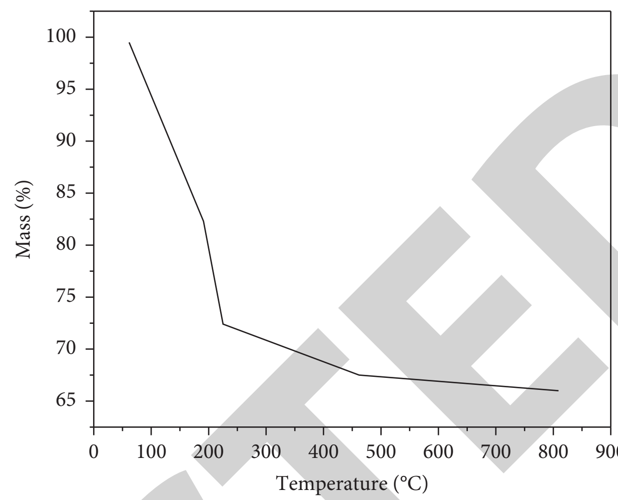

Figure 2: Thermogravimetric curve of the formulation.

From the thermogravimetric curve, it can be seen that the temperature range of severe weight loss of the dried green body is less than 400°C [9]. The water part contained in the hydrated glass material exists in the grid structure in the form of free water and can move back and forth in the gaps of the grid. Some are present in the form of silanol groups (Si-OH). Since there is no chemical change such as oxidative decomposition in the whole system, the whole heating process is the process of losing free water and hydroxyl groups in the system. Before 105°C, the green body first lost about 15 wt.% of free water. Within 105°C–150°C, the crystal water in the system is vigorously removed. Within 150°C–400°C, the weight loss rate of the green body is relatively severe. After 400°C, the weight loss of the dried green body becomes slow. Through multiple experiments, the foaming temperature was determined to be 400°C, the heating rate was 3°C/min, and the holding time was 30 min. At this time, the expansion ratio of the material is larger, the foaming is more uniform, and the obtained low-temperature foaming heat treatment sample is easier to demould, which is convenient for subsequent high-temperature dehydroxylation heat treatment [21].

4.2.2. Material Composition and Structural Analysis. Figure 3 shows Na_2O-B_2O_3-SiO_2 series low-temperature foam glass. Crystalline mineral powder raw material fly ash and formula is mixed sol with fly ash content of 20 wt.% and boric acid content of 2 wt.%. After the low-temperature foaming heat treatment was completed at 400°C for 30 min, the high-temperature dehydroxylation heat treatment was completed at 500°C for 3 h. The X-ray diffraction analysis of the obtained low-temperature foam glass-ceramic composite is shown in Figure 3 [22].

It can be seen that the low-temperature foam glass-ceramic composite is composed of two phases: crystalline phase and amorphous glass phase. The main crystal phase is quartz, and the Na_2O-B_2O_3-SiO_2 low-temperature foam glass is composed of glass phases. Crystalline mineral

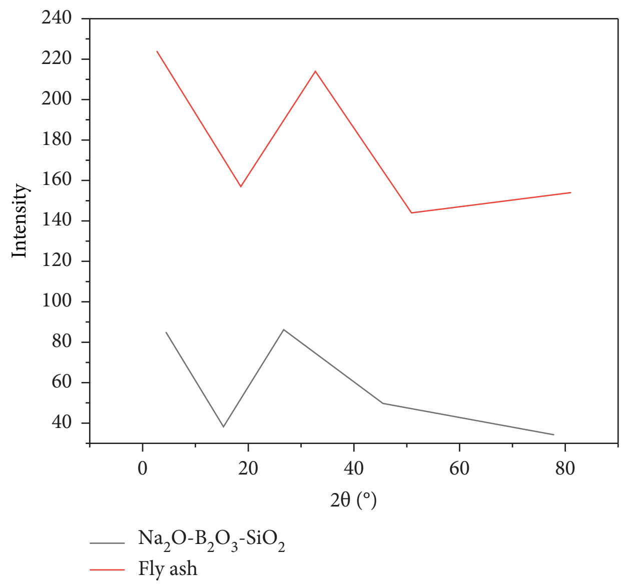

Figure 3: XRD phase analysis of low-temperature foam glass-ceramic composites.

powder fly ash is mainly composed of quartz crystal phase and glass phase [23].

4.3. Exploration and Optimization of Various Process Parameters in Material Preparation

4.3.1. Determination of Glass Matrix Modifier Doping. A series of experiments were carried out using sodium water glass with a modulus of 2.33 and a solid content of 47.22 wt.% as the raw material and boric acid as the glass matrix modifier. The specific raw material composition in formulas 1–4 is shown in Table 1.

According to the ratio of each formula, the boric acid was introduced into sodium water glass by high-speed shearing (4000 rpm/min for 30 min) by colloidal chemical method. A series of hydrated glass sols with different amounts of boric acid were obtained without gelling. It is directly poured into the alumina ceramic crucible and is heated to 300°C. The furnace is according to the heating rate of 39°C/min under free foaming and kept for 30 min. After cooling to room temperature in the furnace, the mold is released to obtain low-temperature foam glass materials with different amounts of boric acid [24]. The influence of the amount of boric acid added on the foaming characteristics and water resistance of Na_2O-B_2O_3-SiO_2-H_2O hydrated glass sol was mainly investigated. Therefore, this series of samples were demolded and cut into pieces for relevant performance tests. The results are shown in Table 2.

The "—"in the above table indicates that the low-temperature foam glass material with the external content of boric acid dissolves or becomes soft after being soaked in warm water at 70°C for 2 hours and loses the corresponding strength, so its weight loss rate and softening coefficient cannot be measured. It can be seen from the table that, with the increase of boric acid content, the bulk density of the

Table 1: Experimental formulas of different boric acid content.

Formula number	Sodium water glass (g)	Boric acid (g)
1	100	2
2	100	3
3	100	4
4	100	5

Table 2: The effect of boric acid doping on the properties of low-temperature foam glass.

Performance	Bulk density	Weight loss rate	Softening factor
1	44	—	—
2	53	—	0.4
3	64	—	0.47

sample gradually increases, and the water resistance improves to a certain extent. This is because the $[BO_4]$ tetrahedron with negative potential is formed after the introduction of the acid into the water glass, and the $[BO_4]$ tetrahedron replaces part of the original $[SiO_4]$ tetrahedron. This further modifies the binary glass system, increasing the integrity of the glass network and improving the water resistance of the material. At the same time, the viscosity of the sol increases, so that the foaming ratio decreases and the bulk density gradually increases. The bulk density of the material does not change much when the external doping amount of succinic acid is 3 wt.% and above. Therefore, the material properties and raw material cost are considered comprehensively: ① When boric acid is introduced into the water glass system, with the increase of the external content of boric acid, the water glass is easier to gel, which will increase the shearing time and the complexity of the process. ② The introduction of boric acid greatly increases the raw material cost of the material. ③ Considering that the subsequent introduction of crystalline mineral powder will also improve the material properties to a certain extent, 2 wt.% boric acid is finally selected.

4.3.2. Effect of Glass Matrix Modifier Types on Material Performance. The experiments were carried out with sodium water glass (modulus of 2.33 and solid content of 47.22 wt.%), boric acid, calcium bentonite, and fly ash (particle size < 25 um). The experimental formulas are shown in Table 3. The effects of glass matrix modifier types on the properties of low-temperature foam glass-ceramic composites were investigated. In the previous section, boric acid was used as the modifier of the glass matrix, and the influence of different dosages of boric acid on the properties of the material was explored, and finally the dosage of 2 wt.% boric acid was optimized. This section explores the effects of different types of glass matrix modifiers and their ratios on material properties.

According to the formulas shown in the table and the experimental procedure shown in Figure 3, a series of mixed sols with different formulas were prepared. According to the heating rate of 3°C/min, with warming up to 400°C and incubation for 30 min, low-temperature foaming heat

Table 3: Experimental formula.

Formula number	Sodium water glass (g)	Fly ash (g)	Boric acid (g)	Calcium bentonite (g)
S1	100	15	0	0
S2	100	15	1	2
S3	100	15	2	1

treatment was carried out. After cooling to room temperature with the furnace, the mold was released, it was directly placed in a furnace at 500°C for 3 h for high temperature dehydroxylation heat treatment. After cooling, it was cut into regular shapes and characterized and tested. The results are as follows.

4.3.3. Bulk Density, Compressive Strength, and Thermal Conductivity. The mixed sol of four groups of proportions completes the low-temperature foaming heat treatment through 400°C of insulation for 30 min and then at 500°C of insulation for 3 h. The bulk density, compressive strength, and thermal conductivity of the low-temperature foam glass-ceramic composite obtained after high-temperature dehydroxylation heat treatment are shown in Figure 4.

It can be seen from the variation law of bulk density that the addition of boric acid reduces the bulk density of the composite material; that is, the foaming performance of the mixed sol is better. After the addition of boric acid, B element captures free oxygen to form a negatively charged $[BO_4]$ tetrahedron, which firmly attracts Na^+ and H^+ in the system, making the original Na_2O-SiO_2 binary system structure tend to be dense, and the viscosity of the mixed sol increases. In the process of low-temperature foaming heat treatment, the uniformity of foaming is improved. However, according to the experimental results, at this stage, the bulk density of the low-temperature foaming heat-treated samples obtained from the two formulations is not much different. The difference occurs in the high-temperature dehydroxylation heat treatment stage. After 3 h of heat preservation at a high temperature of 500°C, the low-temperature foaming sample obtained by the formula has uneven distribution of pores and weak skeleton strength, so volume shrinkage occurs, resulting in an increase in bulk density. The addition of calcium-based bentonite affects the foaming performance of the mixed sol during the low-temperature foaming heat treatment process; that is, the expansion ratio is reduced, resulting in a larger bulk density of the material. The compound modification of boric acid and calcium-based bentonite balances the advantages and disadvantages of both. On the one hand, the low-temperature foaming characteristics are guaranteed. On the other hand, the cost of raw materials is further reduced.

The following can be seen from the variation law of thermal conductivity: after four proportions of samples are processed, the obtained foam glass-ceramic composite thermal insulation material has good thermal insulation performance, and the thermal conductivity is about 0.06 W/(m·K). When boric acid is added, the foaming performance of the material is improved, the pore distribution is uniform,

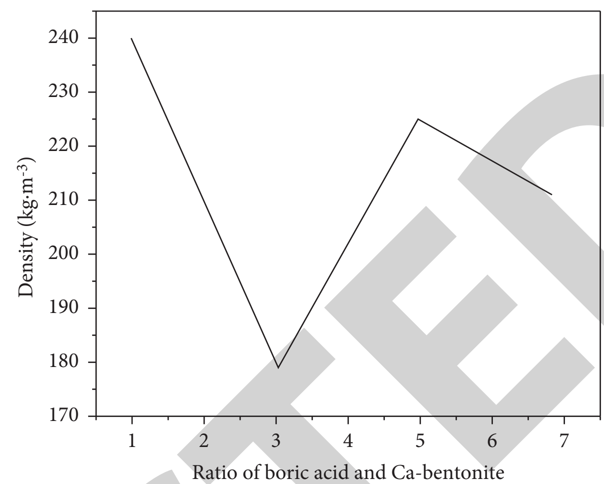

Figure 4: Variation diagram of low-temperature foam glass-ceramic composites obtained with different ratios of boric acid and Ca-bentonite.

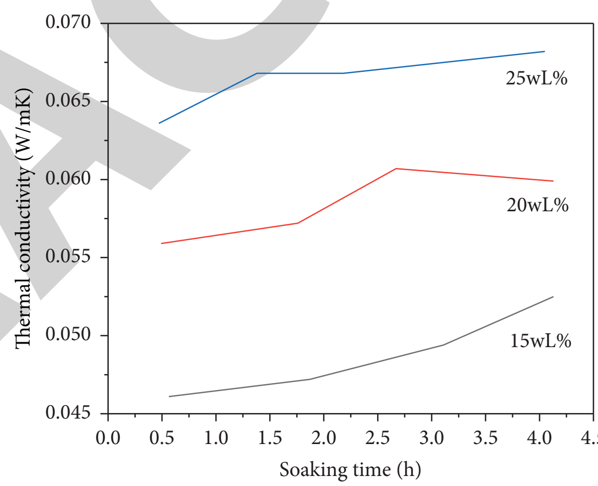

Figure 5: Effect of different soaking time on thermal conductivity of low-temperature foam glass-ceramic composites obtained by different formulations.

and the thermal conductivity is the lowest among the four formulations, only 0.055 W/(m·K). It can be seen that although the addition of calcium bentonite improves the mechanical properties and water resistance of the material, it still affects the expansion ratio of the material, which increases the bulk density and reduces the thermal insulation performance.

4.3.4. Thermal Conductivity. The thermal conductivity test results of the formulations are shown in Figure 5.

With the extension of heat treatment time, the volume shrinkage rate of the material gradually increases, and the internal pores collapse or disappear after being squeezed, the porosity of the material was reduced to a certain extent, and the thermal conductivity increased continuously. It can be seen that, within 2 h of heat treatment, the thermal conductivity of the material did not change much, and the thermal conductivity of each formulation was below 0.06 W/(m·K). When the heat treatment time was 3-4 h, the thermal conductivity of the material increased significantly; in particular, the thermal conductivity of the samples with high fly ash content exceeded 0.06 W/(m·K) and even approached 0.07 W/(m·K). Therefore, it is best to heat the board for about 2 h.

5. Conclusion

In this paper, the preparation of hydrated glass matrix, the introduction of crystalline mineral powder fly ash, and the secondary heat treatment were completed by using colloidal chemical method and using sodium water glass, boric acid, calcium bentonite, and fly ash as the main raw materials. Thus, a low-temperature foam glass-ceramic composite material with lightweight, high strength, good water resistance, and low cost is prepared. At the same time, the process parameters such as dehydroxylation heat treatment temperature and holding time involved in the preparation process were explored and optimized. The conclusions are as follows:

(1) By using the colloidal chemical method, the Na_2O-B_2O_3-Al_2O_3-SiO_2-H_2O system hydrated glass sol is used as the matrix, and the crystalline phase mineral powder fly ash is externally mixed. A low-temperature foam glass-ceramic composite material was prepared through low-temperature foaming heat treatment and high-temperature dehydroxylation heat treatment.

(2) The modification mechanism of crystalline mineral powder fly ash was explored. First, fly ash mainly contains two parts: crystalline phase and amorphous phase. After the fly ash was added, an alkali-aggregate reaction occurred with the water glass. The amorphous component and the glass matrix were dissolved in one phase, and the crystal grains were also reduced in size due to the reaction of alkali aggregates and were evenly distributed in the pore walls. By overlapping each other, they played a certain role of skeleton support, which reduced the problems of hydroxyl content reduction and volume shrinkage during the high-temperature dehydroxylation heat treatment of the material to a certain extent. At the same time, many microcracks were formed on the interface between these grains and the glass phase, which buffered the deformation caused by external force and increased the mechanical strength of the material. Second, after the fly ash was added, the alkali aggregate reacted to form $[AlO_4]$ tetrahedron and $[SiO_4]$ tetrahedron. These tetrahedrons formed long silicate chains through bonding, which improved the integrity and tightness of the glass system and improved the water resistance of the material. Third, after the addition of fly ash, the viscosity of the mixed sol was increased, which affected the foaming of the mixed sol and changed the performance parameters such as the bulk density of the material. With the increase of fly ash content, the modification effect was stronger.

(3) The microstructure of the prepared material was characterized, and a schematic diagram of the structure of the low-temperature foam glass-ceramic composite material was obtained. From the XRD and TEM test results of the composite material, it can be seen that the material is composed of glass phase and crystal phase. The main crystal phase is the quartz crystal phase, wherein the crystal grains of 5–10 nm are uniformly dispersed in the glass matrix.

(4) The exploration and optimization of process parameters were completed, and the optimal raw material ratio and heat treatment system were obtained. For the effects of glass matrix modifier, the content and proportion of boric acid and calcium bentonite, the time of high-temperature dehydroxylation heat treatment, and the content of fly ash on the microstructure and physical and chemical properties of the material were explored. With the prolongation of the holding time of high-temperature dehydroxylation heat treatment, the content of hydroxyl group in the system is continuously reduced, and the water resistance of the material is continuously improved. At the same time, the softening degree of the green body is intensified, the volume shrinkage is gradually increased, the bulk density and thermal conductivity of the material are increased, and the mechanical strength is increased. Finally, when the external content of fly ash is 20 wt.% or more, the external content of boric acid is 1-2 wt.%, and the calcium-based bentonite is 1 wt.%, the mixed sol is subjected to low-temperature foaming heat treatment at 400°C for 30 min and then 500°C high-temperature dehydroxylation heat treatment for 2 hours or more, the bulk density of the obtained low-temperature foam glass-ceramic composite material is 192–256 kg/m^2, the compressive strength is 0.44–0.63 MPa, the weight loss rate is 5.1–7.8 wt.%, the softening coefficient is 0.85–0.95, and the thermal conductivity is 0.056–0.064 W/(m·K).

Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

Conflicts of Interest

The author declares that there are no conflicts of interest.

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