Experimental Study on Preparation of Coal Gangue Magnetic Glass-Ceramics by Direct Sintering Method

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Abstract: Coal gangue is the waste generated during coal mining and processing. This paper aims to efficiently utilize coal gangue as a resource and reduce environmental load. Using coal gangue as the main raw material and copper tailings as auxiliary materials, magnetic glass-ceramics is prepared by direct sintering method. We examine the effects of different raw material ratios, sintering temperatures, and holding times on the structure and properties of magnetic glass-ceramics. The results show that when the coal gangue content is MGS-60wt%, the sintering temperature is 1180 ℃, and the insulation time is 50 minutes, the water absorption rate of magnetic glass-ceramics is 0.0028%, the volume density is 1.269g/cm3, and the magnetization strength is 0.533Gs. As the sintering temperature gradually increases and the holding time increases, its volume density shows a decreasing trend. The magnetization strength of magnetic glass-ceramics belongs to the weak magnetic range, with the lowest magnetization strength of 0.1Gs.

Keywords: Glass-Ceramics; Coal Gangue; Copper Tailings; Direct Sintering Method

1. Introduction

The production and processing of coal inevitably result in corresponding solid waste, with coal gangue being one such byproduct, accounting for approximately 20%. China stands as the world's largest producer of coal gangue [1]. As industrial advancements continue, the demand for high-performance refractory materials and construction supplies grows, posing the need to better utilize coal gangue as a solid waste resource. This paper aims to efficiently harness coal gangue, reduce environmental burdens, and enhance resource utilization. By primarily utilizing coal gangue, supplemented with copper tailings, and employing a direct agglomeration method to produce magnetic glass-ceramics, the study seeks to elevate its value, diminish environmental pollution, and bolster resource recycling efforts.

Due to limited technology and environmental awareness in the past, a substantial amount of abandoned coal gangue was not handled properly. This irresponsible practice led to unrealized potential of these valuable resources, thereby posing significant challenges and difficulties to society and the environment [2]. As a futuristic composite material, glass-ceramics is highly acclaimed. This involves the process of heat treating specific base glass components at a certain temperature, which encourages crystallization. During this process, the base glass uniformly generates a large number of tiny crystals, resulting in the formation of multi-phase composite material comprising glass and nanocrystals, also known as glass-ceramics [3]. Utilizing coal gangue for the production of glass-ceramics represents an innovative technological approach that capitalizes on the high thermal stability and chemical consistency of coal gangue. This transformation of coal gangue turns it into a material of glass-ceramics, equipped with excellent physical properties and promising applications. In the process of manufacturing glass-ceramics, Chen [4] used phosphorus slag and coal gangue as raw materials to achieve an efficient and cost-effective production process. In addition, he studied the effect of different types of phosphorus slag and coal gangue on the melting efficiency and crystal formation capability of glass-ceramics, and finally came up with the optimal ratio of phosphorus slag and coal gangue. Meanwhile, Luo & Zhang [5] and others utilized the direct sintering method to produce glass-ceramics, with coal gangue as the primary raw material and CaO as an aid. They explored the impact of heat treatment on

the physical and chemical properties of the glass-ceramics phase structure, water absorption, volume density and other physical properties. This experiment provided an effective method for the utilization of coal gangue resources. Dang [6] and his team used coal gangue and clay, and discovered that the resulting glass-ceramics was light in weight with high resistance to impact. To achieve optimal mechanical performance, 75% of the coal gangue was matched with 3/1 clay. After continuous research, it was discovered that a sintering temperature of 1,370℃ was optimal and more conducive to the experiment. Xiao [7] and his team successfully prepared high-purity magnetic glass-ceramics target materials, mainly composed of BaFe12O19, for the manufacture of integrated circuit films (ICFs) using a unique melting technology. They employed a four-factor and four-level orthogonal experiment design to evaluate and analyze the material's properties. Eventually, they determined the optimal heat treatment scheme for this material and applied it in practical production. Wang [8] and his team explored the effect of heat treatment on the structure and mechanical properties of glass-ceramics, and found that the material properties were determined by the heat treatment process. After processing at 760℃ for 3 hours and 980 ℃ for 1 hour, glass-ceramics exhibited excellent properties, with a bending strength of over 230MPa which remained stable. Chen [9] and his colleagues evaluated the utilization of various coal mine products at different stages by combining the widespread use of surface soil, fly ash and coal gangue in open-pit mining, and proposed some practical schemes using computer mediation. This provided a solid theoretical basis for promoting the utilization of fly ash, coal gangue and surface soil. Liu [10] and his team outlined the preparation methods and characteristics of ferromagnetic glass-ceramics, focusing on the current research, application status and future development potential.

This paper focuses on the preparation of magnetic glass-ceramics using coal gangue as the primary raw material and copper tailings as the auxiliary material through direct sintering. The effects of different raw material ratios, sintering temperature, and holding time on the structure and properties of magnetic glass-ceramics were investigated. This study provides a theoretical reference for the utilization of other solid waste resources.

2. Experimental Materials and Procedures

2.1 Experimental Materials

In this experiment, coal gangue powder with a particle size of 200 mesh was used.

Chemical composition analysis of coal gangue is crucial for environmental management and resource recovery. As shown in Table 1, SiO_2 is the main component of coal gangue, and its high content indicates that coal gangue is rich in silicon. In addition, it also contains chemical components such as Al_2O_3, Fe_2O_3, CaO, etc. These analytical data of chemical composition provide important information for determining the chemical properties of coal gangue, as well as for its potential applications and treatment.

Table 1. Chemical Composition Analysis (wt%) of Coal Gangue

Compositions	SiO_2	Al_2O_3	Fe_2O_3	CaO	TiO_2	K_2O	SO_3	P_2O_5	SrO
Content	42.01	29.16	9.71	5.44	2.88	1.85	1.25	0.44	0.31
Compositions	BaO	ZrO_2	MgO	Rb2O	Na_2O	MnO	ZnO	NiO	**Loss on ignition**
Content	0.19	0.19	0.16	0.10	0.09	0.06	0.05	0.03	6.08

In Figure 1, the X-ray diffraction pattern of the coal gangue sample was characterized using an X-ray powder diffractometer. As shown in Figure 1, the main components of coal gangue in this experiment are quartz and kaolinite. The peak intensity of quartz is much higher than that of kaolinite, and both peaks appear as sharp peaks. Quartz has a high peak intensity, which indicates that its crystal form is more regular and the shape of the crystal is more symmetrical. During the crystallization process, the high peak intensity of quartz helps to form a more ordered crystal structure, thus benefiting the mechanical properties and thermal stability of the materials. On the other hand, kaolinite may cause irregular crystal shapes and uneven structure during the crystallization process, which could potentially affect the material's performance [11]. Therefore, compared to kaolinite, the high peak intensity of quartz is more advantageous for the formation of crystals and the improvement of material properties during the crystallization process.

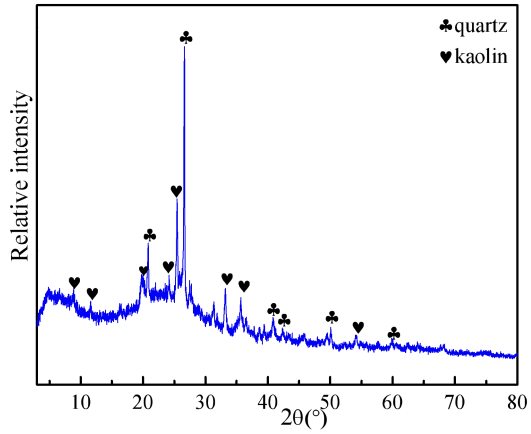


Figure 1. XRD Pattern of Coal Gangue

2.2 Experimental Procedures

According to the experimental plan, three different coal gangue and copper tailings samples with different concentration ratios were weighed using an electronic analytical balance. The total sample weight was 3g. The sample with 80% coal gangue and 20% copper tailings was labeled as MGS-80wt%, the sample with 70% coal gangue and 30% copper tailings was labeled as MGS-70wt%, and the sample with 60% coal gangue and 40% copper tailings was labeled as MGS-60wt%. After weighing the samples, they were ground thoroughly in a mortar to adjust the particle size to the optimal state. The powder was then poured into a crucible for the next step and 3 mL of PVA solution was added using a pipette to ensure that the powder was well mixed with the PVA. Next, the mixture was gently stirred using a glass rod until no grains were attached to the container or the rod, indicating that the powder was well combined with the PVA. Finally, the granulation process was completed. The well-stirred raw materials were poured into the powder press mold and the pressure was set to 21 MPa. After waiting for a few minutes, the glass-ceramics billet was taken out. After standing for 24 hours, it was placed in a box furnace and sintered according to the predetermined sintering conditions. The sintering temperature was set to 1,100℃, 1,120℃, 1,140℃, 1,160℃, and 1,180℃, and the holding time was set to 10min, 20min, 30min, 40min, and 50min, respectively. After the sintering was completed, the furnace door was opened, and the internal temperature was gradually lowered to room temperature. The sintered samples were then taken out, labeled, and recorded.

2.3 Sample of Magnetic Glass-Ceramics Prepared from Coal Gangue

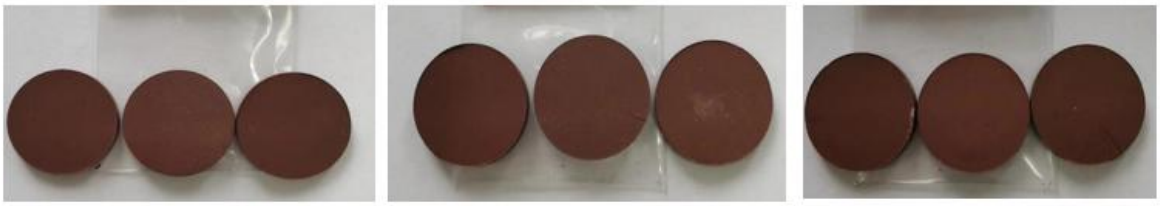
2.3.1 Effects of different sintering temperatures on the morphology of magnetic glass-ceramics

Figures 2-4 display the morphology of magnetic glass-ceramics samples processed by direct sintering. The sintering temperatures were 1,100℃, 1,140℃, and 1,180℃, respectively, and the coal gangue content (MGS-80wt%, MGS-70wt%, MGS-60wt%) and holding time were kept constant at 50 minutes. Three parallel samples were prepared for each group. Under the conditions of sintering temperature at 1,100℃, the color of the sintered samples was brick red. The surface of all three samples with different coal gangue contents appeared relatively flat and smooth, with little color difference between them. Under the conditions of sintering temperature at 1,140℃, the color of the samples changed from brick red to reddish brown. The surface of the samples had some small particles and unevenness. The second parallel sample of MGS-60wt% had a slight bulge on the surface due to incomplete mixing during the preparation of the glass-ceramics billet, resulting in partial bulges in the sintered sample. The third parallel sample had a slight crack due to improper use of the mold during specimen compression. Under the conditions of sintering temperature at 1,180℃, the color of the samples changed significantly, appearing dark brown. The MGS-60wt% sample had some microcracks, and the overall surface of the samples appeared more granular than at the lower temperatures. It can be concluded that as the sintering temperature increases, the color of the samples gradually darkens. When the sintering temperature reaches a certain value, cracks may occur on the surface of the samples, which may not be as smooth as at lower temperatures.

2.3.2 Effects of different sintering temperatures on the morphology of magnetic glass-ceramics

Figures 5-7 show the sintered samples obtained with different coal gangue contents at a sintering temperature of 1,180℃ for sintering times of 10, 30, and 50 minutes, respectively. It is observed that the sintered samples all had a circular shape with little variation in morphology. The color of the

samples gradually changed from reddish-brown to dark-brown as the sintering time increased, and there were no significant color differences among the three different coal gangue content samples when the holding time was 10 min. However, when the holding time was extended to 30 minutes, some small particle-like substances could be observed on the surface of the samples, and there was no significant change in the color of the samples, although it was observed that the surface color of the sintered MGS-80wt% sample was slightly lighter than the other two samples. When the holding time reached 50 minutes, the color of the samples changed slightly to dark-brown. As the holding time was prolonged, the particles became more visible, and the surface became rough. It was observed that the first parallel sample of MGS-60wt% and the third parallel sample of MGS-70wt% expanded slightly when the holding time was 30 min and 50 min, which was caused by incomplete mixing during the preparation of magnetic glass-ceramics billets that led to partial expansion of the sintered samples.



(a) MGS-60wt% (b)MGS-70wt% (c)MGS-80wt%

Figure 2. Morphology of different coal gangue contents (1,100℃-50min)

(a)MGS-60wt% (b)MGS-70wt% (c)MGS-80wt%

Figure 3. Morphology of different coal gangue contents (1,140℃-50min)

(a)MGS-60wt% (b)MGS-70wt% (c)MGS-80wt%

Figure 4. Morphology of different coal gangue contents (1,180℃-50min)

(a)MGS-60wt% (b)MGS-70wt% (c)MGS-80wt%

Figure 5. Morphology of different coal gangue contents (1,180℃-10min)

(a)MGS-60wt% (b)MGS-70wt% (c)MGS-80wt%

Figure 6. Morphology of different coal gangue contents (1,180℃-30min)

(a)MGS-60wt% (b)MGS-70wt% (c)MGS-80wt%

Figure 7. Morphology of different coal gangue contents (1,180℃-50min)

2.4 Sample Performance Testing

(1) Determination of water absorption and bulk density: The magnetic glass-ceramics samples were dried in a vacuum drying oven until completely dry and cooled to room temperature. The mass of the dried sample was measured and recorded as M0a. Then, the sample was placed in a self-sealing bag and a certain amount of distilled water was added to the bag so that the sample was completely immersed in the water. After soaking for 24 hours, the sample was removed and the surface moisture was wiped dry with a tissue. The weight of the sample was measured again and recorded as M0b. The distilled water was poured into a wire basket and the entire wire basket was immersed in the distilled water. Finally, the magnetic glass-ceramics sample was placed into the wire basket, and its weight (M0c) in the water body was recorded. The calculation formulas were as follows:

$$§ = \frac{M_{0b}-M_{0a}}{M_{0a}} \times 100\% \quad (1)$$

$$\rho_a = \frac{M_{0a}}{M_{0b}-M_{0a}} \times \rho_1 \quad (2)$$

Where: § is the water absorption rate, %; M0a is the weight of the sample after drying, g; M0b is the weight of the sample after fully soaking in distilled water for 24 h, g; ρa is the bulk density, g/cm3; ρ1 is the density of distilled water, g/cm3; and M0c is the weight of the sample in water, g.

(2) Measurement of magnetization intensity: The magnetization intensity of the tested samples in this experiment was measured using a handheld digital Tesla meter. Firstly, after the sintering process, the sample was taken out of the box-type atmosphere furnace and placed horizontally on the table after cooling to room temperature. Then, the sensor tip of the handheld digital Tesla meter was touched onto the surface of the sample to obtain the magnetization intensity reading. The measurement results showed that the prepared magnetic glass-ceramics belonged to weak magnetism and met the weak magnetic field standards of the People's Republic of China [12].

3. Results and Analysis Discussion

3.1 Effect of Sintering Temperature on the Properties of Magnetic Glass-Ceramics

3.1.1 Effect of sintering temperature on water absorption of magnetic glass-ceramics

Figure 8 shows the water absorption data of magnetic glass-ceramics samples with different contents sintered at temperatures ranging from 1,100°C to 1,180°C with a holding time of 50 minutes. The plot was generated using Origin software. It can be observed that the water absorption rates of MGS-80wt%, MGS-70wt%, and MGS-60wt% samples generally decrease with the increase of sintering temperature, and the water absorption rates of the three contents are the lowest at 1,180°C. The water absorption rate of MGS-80wt% is 0.0027%, while that of

MGS-70wt% and MGS-60wt% are 0.0022% and 0.0028%, respectively. This trend is due to the better compactness of the samples at higher sintering temperature, and water is less likely to penetrate into the interior of the sample. During the sintering process of coal gangue in the preparation process, the organic matter in the coal gangue is fully burned, which reduces the pore structure of the glass-ceramics and thus reduces its water absorption rate [13]. It can be inferred from the experimental results that the change in water absorption rate is closely related to the sintering temperature, and as the sintering temperature increases, the water absorption rate of the sample decreases.

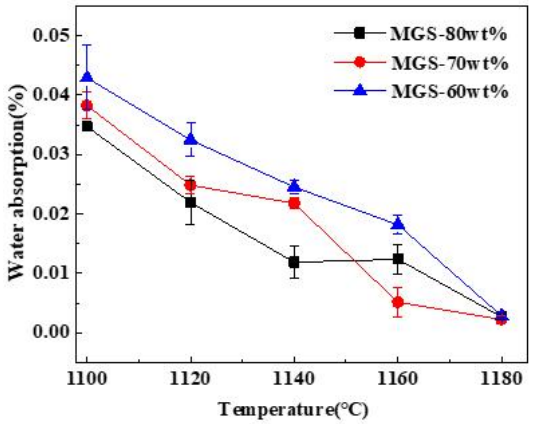


Figure 8. Water Absorption (1,100℃-1,180℃, 50min)

3.1.2 Effects of different sintering temperatures on the volume density of magnetic glass-ceramics

Figure 9 shows the variation of volume density with sintering temperature when the holding time is fixed at 50 minutes. It can be observed that the volume density of the three samples with different contents all exhibit a decreasing trend, and their volume densities reach the minimum at 1,180°C. As the sintering temperature increases, the liquid phase content formed by the high-temperature melting of quartz and amorphous glass in the sample increases, and the viscosity of the glass decreases due to the weakening of molecular forces. The sufficiently melted liquid phase undergoes viscous flow, gradually merging the small pores between the raw material particles to form large pores, and the volume density of the sample decreases [14]. Therefore, it can be known that the volume density of the sample is affected by the sintering temperature, and the higher the sintering temperature, the lower the volume density.

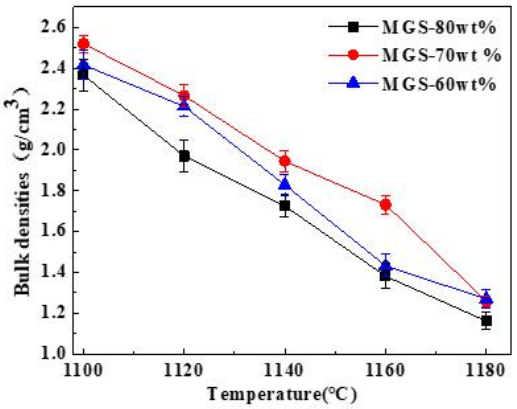


Figure 9. Volume Density (1,100℃-1,180℃, 50min)

3.1.3 Effects of different sintering temperatures on the magnetization of magnetic glass-ceramics

Figure 10 shows the magnetization data of samples sintered at temperatures ranging from 1,100°C to 1,180°C with a holding time of 50 minutes. It can be observed that the magnetization of the sample is related to the content of copper tailings. For MGS-80wt%, the magnetization of the sample shows no regular change, and as the content of copper tailings gradually increases, the magnetization of the sample shows an overall increasing trend. MGS-60wt% exhibits an increasing trend, reaching a peak value of 0.533 Gs at 1,180°C. Copper tailings are rich in iron minerals, such as magnetite and hematite [15]. Therefore, samples containing more iron minerals in the copper tailings may exhibit higher magnetization.

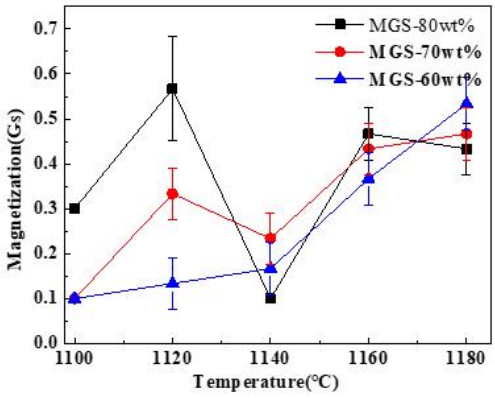


Figure 10. Magnetization (1,100℃-1,180℃, 50min)

3.2 Effects of Holding Time on the Properties of Magnetic Glass-Ceramics

3.2.1 Effects of different holding time on the water absorption of magnetic glass-ceramics

Figure 11 shows the changes in water absorption of samples sintered at the same temperature (1,180°C) with different holding times. It can be observed that the water absorption of MGS-80wt% fluctuates greatly, and reaches the lowest value of 0.0019% at a holding time of 40 minutes. MGS-70wt% and MGS-60wt% exhibit a decreasing trend in the time interval of 10 to 40 minutes, and both reach the lowest value at a holding time of 40 minutes, which are 0.0017% and 0.0019%, respectively.

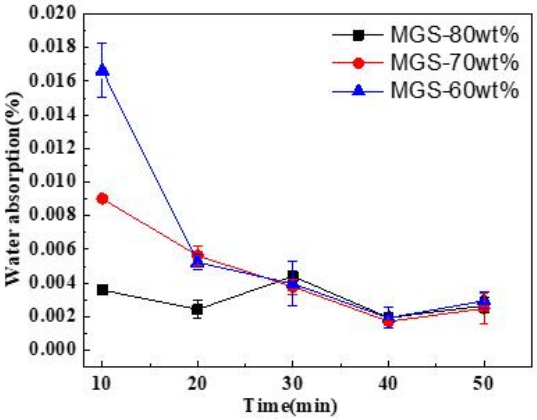


Figure 11. Water Absorption (1,180℃, 10min-50min)

3.2.2 Effects of different holding times on the volume density of magnetic glass-ceramics

Figure 12 shows the changes in volume density of samples sintered at a constant temperature of 1180°C with different holding times (from 10 to 50 minutes). It can be observed from Figure 12 that the volume density of all three samples decreases as the holding time increases. MGS-80wt%, MGS-70wt%, and MGS-60wt% all reach their lowest volume densities at a holding time of 50 minutes, which are 1.162g/cm3, 1.259g/cm3, and 1.326g/cm3, respectively.

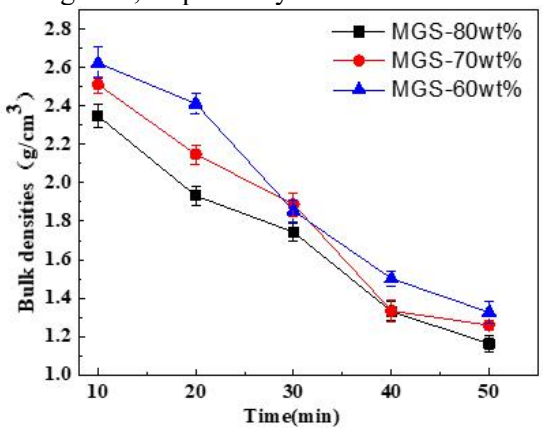


Figure 12. Volume Density (1,180℃, 10min-50min)

3.2.3 Effects of different holding times on the magnetization of magnetic glass-ceramics

Figure 13 illustrates the changes in magnetization of samples sintered at a constant temperature of 1,180°C with different holding times. It can be observed that MGS-80wt% exhibits irregular fluctuations and reaches its highest value of 0.433Gs at a holding time of 50 minutes. MGS-70wt% shows an overall increasing trend, with the fastest increase in the time interval of 20 to 40 minutes, and reaches its highest value of 0.5Gs at a holding time of 50 minutes. MGS-60wt% exhibits a relatively gradual increase and reaches its highest value of 0.533Gs at a holding time of 50 minutes. The differences in the trends of MGS-80wt% compared to MGS-70wt% and MGS-60wt% can be attributed to the difference in copper tailings content, where a higher content of copper tailings leads to a more regular increase in magnetization.

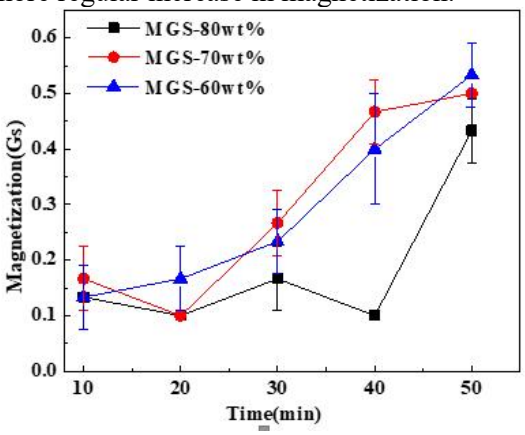
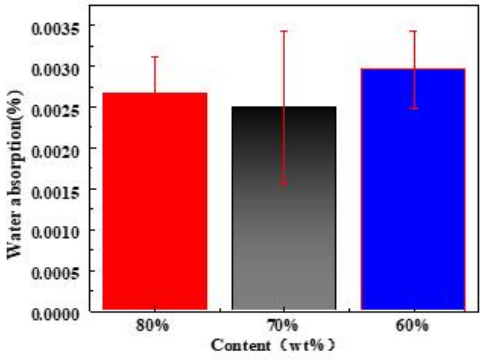


Figure 13. Magnetization (1,180℃, 10min-50min)

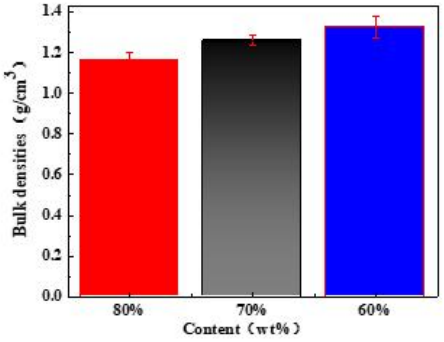
3.2.4 Effects of different contents on the properties of magnetic glass-ceramics

Figure 14 shows the test values of the properties of samples sintered at the same temperature (1,180°C) and with the same holding time (50 minutes) but with different coal gangue contents. It can be observed that at 1,180°C and 50 minutes holding time, MGS-70wt% has the lowest water absorption among the three samples, with MGS-80wt% having slightly lower water absorption compared to MGS-60wt%. The volume density increases with the decrease in coal gangue content, where MGS-80wt% has the lowest volume density of 1.162g/cm3. The magnetization strength increases with the increase in copper tailings content, with MGS-60wt% having the highest magnetization value of 0.533Gs when

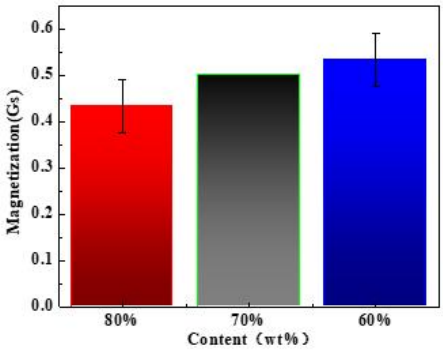
the copper tailings content is the highest. Considering the priority of magnetization strength in this study on the preparation of magnetic glass-ceramics with low water absorption and low volume density, it can be concluded from the experimental data that when sintered at 1,180°C for 50 minutes, MGS-60wt% exhibited the highest magnetization strength of 0.533Gs. Therefore, the optimal process parameters are: coal gangue content of MGS-60wt%, sintering temperature of 1180°C, and holding time of 50 minutes, resulting in a water absorption of 0.0028%, a volume density of 1.269 g/cm3, and a magnetization strength of 0.533Gs.



(a)Water absorption



(b)Volume density



(c)Magnetization

Figure 14. Properties of sintered samples (1,180℃-50min)

4. Conclusions

(1) Coal gangue is primarily composed of SiO2, indicating a significant presence of siliceous materials. Other chemical components, such as Al_2O_3, Fe_2O_3, and CaO, are also present. Since silicon and aluminum are both key components in glass production, coal gangue has potential value in the fabrication of magnetic glass-ceramics.

(2) Quartz and kaolin are the main components in coal gangue, with quartz exhibiting a significantly higher peak intensity compared to kaolin. The high peak intensity of quartz suggests a more regular crystal form and symmetrical shape, facilitating the formation of an ordered crystal structure during the crystallization process. Meanwhile, kaolin may lead to irregular crystal forms and uneven structures, potentially impacting the material's properties. In short, quartz's high peak intensity is preferable over kaolin in facilitating crystal formation and raising material properties during the crystallization process.

(3) Increasing the sintering temperature results in higher liquid phase content from the high temperature melting of quartz and amorphous glass in the sample. This decrease in glass viscosity due to weakening molecular forces causes the molten liquid phase to flow more easily, filling in the gaps between raw material particles, and reducing sample density. A higher sintering temperature leads to better sample compactness, making it harder for water to penetrate the internal structure and reducing the sample's water absorption rate. Finally, copper tailings with more iron minerals, such as magnetite and maghemite, may exhibit higher magnetization strengths.

(4) Optimal conditions for manufacturing magnetic glass-ceramics include a coal gangue content of MGS-60wt%, a sintering temperature of 1,180°C, and a holding time of 50 minutes. Under these conditions, the glass exhibits a water absorption rate of 0.0028%, a volume density of 1.269g/cm3, and a magnetization strength of 0.533Gs.

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