Study on the preparation and influence of alkali metal oxides R$_2$O on properties of microfiber glass with high water resistance

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Abstract: The microfiber glass with high water resistant was prepared by the centrifugation with synchronous water-repellent atomization spray. The influence of the alkali metal oxide Na$_2$O/(Na$_2$O+K$_2$O) ratio on the fiber forming ability, chemical stability, thermal and acoustical insulation properties of the Si-Ca-Mg-Al-Fe-B multi-glass fiber system was studied by combing with the “mixed alkali effect”. It was found that when the ratio Na$_2$O/(Na$_2$O+K$_2$O)=0.97, the fiber forming ability and chemical stability of the prepared microfiber glass were optimized. The transmission loss and the sound absorption coefficient are distributed at 2000Hz at a maximum of 9.1dB and 0.88. Therefore, under this paper preparation conditions the optimal Na$_2$O and K$_2$O distribution ratio of Si-Ca-Mg-Al-Fe-B multi-glass fiber system is 16wt% and 0.5wt%, respectively. Each prepared fiber is tightly connected together by the surface treatment of the water repellent to form a stable fiber network structure, demonstrating the feasibility of producing high water resistant microfiber glass by the centrifugation with synchronous water-repellent atomization spray in practical production applications.

1. Introduction

Energy is the material basis for human survival and an important support and key strategic material for the country’s economic development. At present, China is in a rapid stage of urbanization development so that the huge energy is demanded. Furthermore, this demand will become more urgent in the near future. Therefore, the development of green intelligent buildings towards smart ecological cities is the inevitable direction for solving the energy crisis, resulting in that the development of high-performance green building materials is becoming the core of development [1-3].

Microfiber glass is a new type of green building material, which possesses the excellent thermal insulation performance, non-combustible, light weight, electrical insulation and corrosion resistance to provide people a comfortable and safe living environment and reduce the overall energy consumption of building construction and operation by reducing CO$_2$ emissions [4-5]. In the composition system of microfiber glass, alkali metal oxide R$_2$O corresponding to Na$_2$O and K$_2$O has a great influence on the physical and chemical properties of microfiber glass [6], but the study on the effect of alkali metal oxide R$_2$O on the properties of microfiber glass by combing with the “mixed alkali effect” are rare
In this paper, the microfiber glass was prepared by the centrifugation with synchronous water-repellent atomization spray. The effect of the alkali metal oxide Na$_2$O/(Na$_2$O+K$_2$O) ratio on the fiber forming ability, chemical stability, thermal insulation performance and sound insulation performance of Si-Ca-Mg-Al-Fe-B microfiber glass system was researched, which has a guiding help for further improving the performance of microfiber glass used in the green building materials.

2. Experimental method

2.1 Sample preparation

The microfiber glass with high water resistant mentioned in this paper is prepared by the simultaneous process of centrifugation and atomization spray. First, according to the chemical designed composition of the microfiber glass shown in Table 1, selecting the appropriate amount of quartz sand, soda ash, potassium feldspar, albite, calcite, borax, dolomite, barium carbonate and zinc oxide raw materials, and then the raw material mixture was poured into the furnace with the temperature of 1300-1450°C for the high-temperature calcination in order to remove bubbles and impurities and melt the mixture into a uniform transparent glass liquid. Then the glass liquid flowed through the nickel-cobalt alloy bushing into the centrifuge, and the inflowing glass liquid (850-1000°C) was centrifuged by high-speed rotation spinner with the small holes ($ø=0.8\text{mm}$), resulting in taking out of microfiber glass with an average fiber diameter of $ø=2.5\mu\text{m}$. Meanwhile, the 2.5wt% hydroxy silicone oil water-repellent agent was sprayed uniformly on the surface of the prepared microfiber glass during the falling collection process. The surface-treated fiber was evenly adsorbed and laid on the mesh belt of the fiber collecting machine with a frequency of 32 to 35 Hz negative induced pressure according to a certain fiber arrangement. Finally, the prepared microfiber glass was dried at 150-175°C for 3-4mins to form a high water-resistant microfiber glass wool with an areal density of 2000g/m$^2$.

Table 1 the chemical composition of microfiber glass

| SiO$_2$  | CaO  | MgO | Al$_2$O$_3$ | Fe$_2$O$_3$ | B$_2$O$_3$ | R$_2$O | (Na$_2$O+K$_2$O) |
|----------|------|-----|-------------|-------------|-----------|-------|-----------------|
| 64.5     | 6.2  | 3.0 | 3.2         | 0.13        | 6.5       | X= Na$_2$O/(Na$_2$O+K$_2$O) |
|          |      |     |             |             |           | (X=0.85, 0.88, 0.91, 0.94, 0.97, 1) |

2.2 Test characterization

First, a sample of microfiber glass wool with a size of 50×15mm was taken to weigh and record the value as $m_1$. Then the sample was soaked into a plastic container with 500g of pure water (pH=7±0.1) for 24 hours, and then its weight recording as $m_2$. So the hydrolysis rate $\rho$ of sample can calculated by the following the formula (1),

$$\rho=(m_1-m_2)/m_2\times10^6$$  

Where: $m_1$: weight before soaking (g); $m_2$: weight after soaking (g); $\rho$: rate of hydrolysis (ppm).

The German Netzsch HFM 436 heat flow analyzer was used to test the thermal conductivity $\lambda$ of the prepared microfiber glass sample at room temperature. Before testing, the sample was cut into a 300×300mm square and placed horizontally on the cold plates of analyzer, and the opposite hot plate was pressed down to compress the sample to a certain thickness before starting the test. The coefficient of thermal expansion $\alpha$ of the prepared microfiber glass sample was measured using a German NETZSCH DIL 402C dilatometer. A strip sample with a size of 10×10×50mm was first prepared, and the test temperature was set to 25-300 °C.

The surface and cross-section microstructure of the prepared microfiber glass sample was observed and characterized by Japanese electronic JEOL JSM-6360 scanning electron microscope (SEM). Since the glass sample is not electrically conductive, it is necessary to process the gold spray treatment before the observation.

The acoustic-insulation performance test of prepared microfiber glass sample was carried out by the standing wave tube method. The test system is the sound absorption coefficient of AWA6290T.
transfer function.

3. Results and analysis

3.1 Analyzing fiber forming ability and chemical stability of prepared microfiber glass sample

Table 2 shows the effect of the ratio of alkali metal oxide \( \frac{Na_2O}{(Na_2O+K_2O)} \) on the melting temperature \( T_m \), forming temperature \( T_w \), liquidus temperature \( T_l \), forming temperature interval \( \Delta T \) and hydrolysis performance \( \rho \) of Si-Ca-Mg-Al-Fe-B multi-glass fiber system. It can be seen from Table 2 that as the ratio of the alkali metal oxide \( Na_2O/(Na_2O+K_2O) \) increases, the \( T_m, T_w, T_l \) and \( \rho \) of the prepared microfiber glass sample gradually decrease and \( \Delta T \) gradually increases. When \( Na_2O/(Na_2O+K_2O)=0.97 \), the samples \( T_m, T_w, T_l \) and \( \rho \) obtain the minimum value, respectively. \( \Delta T \) takes the maximum value. When the ratio of alkali metal oxide \( Na_2O/(Na_2O+K_2O) \) further enhances to be 1, corresponding to the alkali metal oxide in the ultrafine glass fiber cotton is all \( Na_2O \), the \( T_m, T_w, T_l \) and \( \rho \) of sample rise again, and \( \Delta T \) appears to fall.

Table 2 the effect of the ratio of alkali metal oxide \( Na_2O/(Na_2O+K_2O) \) on the melting temperature \( T_m \), forming temperature \( T_w \), liquidus temperature \( T_l \), forming temperature interval \( \Delta T \) and the hydrolysis performance \( \rho \) of Si-Ca-Mg-Al-Fe-B multi-glass fiber system

| Sample | \( X=Na_2O/ \) \( (Na_2O+K_2O) \) | \( T_m/°C \) | \( T_w/°C \) | \( T_l/°C \) | \( \Delta T/°C \) | \( \rho/ppm \) |
|--------|-----------------|--------------|--------------|--------------|----------------|----------------|
| 1      | 0.85            | 1452         | 1141         | 1066         | 75             | 1198           |
| 2      | 0.88            | 1432         | 1128         | 1031         | 97             | 1122           |
| 3      | 0.91            | 1415         | 1098         | 986          | 112            | 1058           |
| 4      | 0.94            | 1403         | 1065         | 940          | 125            | 995            |
| 5      | 0.97            | 1382         | 1011         | 869          | 142            | 978            |
| 6      | 1.00            | 1398         | 995          | 873          | 122            | 1047           |

The variation curve of the forming temperature interval \( \Delta T \) and the hydrolysis performance \( \rho \) of the Si-Ca-Mg-Al-Fe-B multi-component glass fiber system with the ratio of the alkali metal oxide \( Na_2O/(Na_2O+K_2O) \) is shown as Fig.1. When \( Na_2O/(Na_2O+K_2O)=0.97 \), corresponding to the mass fractions of \( Na_2O \) and \( K_2O \) as be 16wt% and 0.5wt%, respectively, the forming temperature range reaches the maximum value \( \Delta T_{max}=142°C \), and the hydrolysis performance reaches the minimum value \( \rho_{min}=978ppm \). The forming temperature interval \( \Delta T \) reflects the difficulty in forming the glass fiber, resulting from the wider the forming temperature range, the easier the formation of the microfiber glass. In the process of preparing microfiber glass by the centrifugation and simultaneous atomization spray, since the large contact area and the long contact time between the centrifugal spinner and the glass liquid, when the melting temperature, the forming temperature and the liquid phase temperature are not properly controlled, leading to the increase of crystallization possibility of the glass liquid. Once the glass liquid appears submicron-crystal or full crystal, it is easy to cause clogging for microporous of the centrifugal spinner, which makes it difficult to prepare the microfiber glass by the centrifugation method. At the same time, since the centrifugal spinner is limited by the temperature resistance of the material, centrifugal fiberglass is required to be prepared at the liquidus temperature as low as possible. If the molten temperature of glass system is too high, the centrifugal spinner and the bushing will be eroded by the high temperature molten glass, which will greatly shorten their service life, thereby increasing the forming difficulty and production cost of the preparation of microfiber glass. The smaller \( \rho \) value of the prepared microfiber glass sample, the more stable the chemical properties of the sample. Both \( Na_2O \) and \( K_2O \) are the most common alkali metal oxides in high-silica glass system. The \( Na_2O \) provides the oxygen ion with high polarization for the glass fiber system, which mitigates the competition of \( Si^{4+} \) with metal ions such as \( Ca^{2+}, Mg^{2+} \) and \( Al^{3+} \) for oxygen ions, polarizes bridge oxygen and weakens the silicon-oxygen bond, resulting in loosening glass fiber structure. Therefore, adding \( Na_2O \) can reduce the high temperature viscosity of the glass system, and the chemical stability of the prepared microfiber glass is lowered. Due to the mixed alkali effect, when the \( Na_2O \) is partially replaced by the \( K_2O \) corresponding to the ratio of \( Na_2O/(Na_2O+K_2O) \)
decreasing, the forming temperature interval $\Delta T$ and the hydrolysis performance $\rho$ don’t change in a unidirectional linear relationship, but there is an inverse linear transformation and appears the extreme value. Due to that the $K^+$ radius is larger than $Na^+$ and its field strength is smaller than that of $Na^+$, so the agglomeration capacity for the surrounding non-bridge oxygen ions of $K^+$ is weaker than that of $Na^+$, resulting in that the excessive free oxygen makes the internal network structure destruction ability of the glass fiber system enhanced, the chemical bond is aggravated and the network structure becomes loose when $K_2O$ partial replacement of $Na_2O$ in glass fiber system compared with only pure $Na_2O$. Therefore, when the ratio of $Na_2O/(Na_2O+K_2O)$ is equal to 0.97, the optimum ratio of $Na_2O$ and $K_2O$ in the Si-Ca-Mg-Al-Fe-B multi-glass fiber system is 16 and 0.5wt%, respectively. The fiber forming ability and chemical stability of the prepared microfiber glass sample under the reported test conditions are optimized.

3.2 Analyzing of expansion coefficient and thermal conductivity of ultra-fine glass fiber cotton

Fig.2 is a graph showing the expansion coefficient $\alpha$ and the thermal conductivity $\lambda$ of the Si-Ca-Mg-Al-Fe-B multi-glass fiber system as a function of the ratio of $Na_2O/(Na_2O+K_2O)$. As shown in Fig.2, as the ratio of $Na_2O/(Na_2O+K_2O)$ increases, the $\alpha$ and $\lambda$ of the prepared microfiber glass sample gradually decrease. When $Na_2O/(Na_2O+K_2O)=0.97$ corresponding to the mass fraction of $Na_2O$ and $K_2O$ as 16 and 0.5wt%, the expansion coefficient and thermal conductivity obtain the minimum value $\alpha_{\text{min}}=86.283^\circ\text{C}^{-1}$ and $\lambda_{\text{min}}=0.028\text{W/(m}\cdot\text{K})$, respectively. But when the ratio of $Na_2O/(Na_2O+K_2O)$ further enhance to 1 representing the all alkali metal oxide in multi-glass fiber system as $Na_2O$, the $\alpha$ and $\lambda$ value appear to be increasing.

The main reason for the above phenomenon is that when the $K_2O$ partially replacing part of $Na_2O$ in the glass fiber system, the repulsive force between the pure $Na^+$ is greater than that between $Na^+$ and $K^+$, so that the binding force of $Na^+$ and $K^+$ in the mixed alkali system with the glass fiber network is greater than that of the single alkali metal ions. When the free path of the gas molecule is larger than the pore diameter of microfiber glass wool, the gas molecules are generally in a static state, and most of the gas molecules are adsorbed on the pore walls inside the material and a large number of semi-connected or closed pore networks inside the microfiber glass wool. Meanwhile, the structural factors of microfiber glass wool make it difficult for gas molecules to undergo thermal convection. The layered of microfiber glass facilitates dense overlap and uniform dispersion between fibers and reduces heat convection. In addition, since heat conduction can only be conducted between solids, the thermal conduction can only pass through a small number fiber of each fiber layer due to the layered structure characteristics of microfiber glass wool.

With the decrease of the ratio of $Na_2O/(Na_2O+K_2O)$ corresponding to the alkali $K_2O$ more substituting for $Na_2O$ in fiber glass system, the migration of alkali metal ions is restricted the viscosity of the fiber glass system is limited, resulting in the

![Fig.1 The effect of the ratio of $Na_2O/(Na_2O + K_2O)$ on the forming temperature interval $\Delta T$ and hydrolysis performance $\rho$.](image1)

![Fig.2 The effect of the ratio of $Na_2O/(Na_2O + K_2O)$ on the expansion coefficient $\alpha$ and thermal conductivity $\lambda$.](image2)

continuous crystallization and an increase in thermal conductivity of prepared microfiber glass due to the increased thermal conductivity of the glass fiber skeleton. Therefore, the optimum ratio of $Na_2O$ and $K_2O$ in the Si-Ca-Mg-Al-Fe-B multi-glass fiber system is 16wt% and 0.5wt%. 

4
3.3 Analyzing of sound insulation performance of prepare microfiber glass

Fig. 3 is shown a sound insulation characteristic curve of Si-Ca-Mg-Al-Fe-B multi-component fiber glass system with the ratio of Na$_2$O/(Na$_2$O+K$_2$O). As shown as Fig. 3 (a, b), at the same noise frequency, the comparison of sound transmission loss and the sound absorption coefficient of prepare microfiber glass increase continuously with increasing the Na$_2$O/(Na$_2$O+K$_2$O) ratio. When Na$_2$O/(Na$_2$O+K$_2$O)=0.97 corresponding to the mass fraction of Na$_2$O and K$_2$O as be 16wt% and 0.5wt%, respectively, the sound transmission loss and sound absorption coefficient of the sample are at 2000Hz frequency reach to the maximum values of 9.1 dB and 0.88, respectively. When the ratio of Na$_2$O/(Na$_2$O+K$_2$O) is further increased to 1, that is, the alkali metal oxide in the prepared microfiber glass is all Na$_2$O, the sound transmission loss and the sound absorption coefficient of the sample appear to be declining.

The sound insulation performance of prepared microfiber glass sample mainly depends on the material's own characteristics (surface density, stiffness, damping and other parameters), the internal fiber network structure and the frequency of incident sound waves. Since this paper mainly studies the effect of alkali metal oxide Na$_2$O/(Na$_2$O+K$_2$O) ratio on the sound insulation performance of microfiber glass sample, the sound insulation characteristic curve (stiffness damping control area) of the sample in the low frequency range (≤3000Hz) is selected as the research target. The sound insulation is proportional to the stiffness and increases with the stiffness of the prepared sample.

The accumulation ability of K$^+$ is weak for its surrounding non-bridged oxygen ions, resulting in more free oxygen in the mixed alkali fiber glass system and increasing the internal network structure damage and loose structure, finally causing chemical bond breakage and rigidity reduction for the prepared microfiber glass sample.

When sound waves in a certain frequency are incident on the surface of the prepared sample, the sound waves are divided into two parts to propagate in different directions: one part is reflected on the surface of the prepared sample and the other part is propagated into the inside of the sample. Fig. 3(a) is a partial SEM picture of a sample of prepared microfiber glass for the Si-Ca-Mg-Al-Fe-B multi-glass fiber system when the ratio of Na$_2$O/(Na$_2$O+K$_2$O) is 0.97. It can be seen that the uniform...
atomization sprayed 2.5wt% hydroxy silicone oil water repellent on the sample surface, and each microfiber glass is closely connected by the surface treatment of the water repellent to form a stable fiber network. The incident of lower frequency sound wave doesn’t cause the damage of internal fiber structure of the prepared sample. Fig. 3(b) is a partially enlarged SEM image of the sample shown in Fig. 3(a), the surface-modified microfiber glass are bonded together by the action of the water repellent to improve the stability of the fiber structure in order to ensure a three-dimensional network structure. Therefore, the prepared microfiber glass sample sound transmission loss exceeds 9 dB at a frequency of 2000 Hz.

4. Conclusion
When Na2O/(Na2O+K2O)=0.97, the forming temperature interval ΔT of the prepared microfiber glass sample reaches to the maximum value ΔTmax=142°C; The hydrolysis performance reaches the minimum value ρmin=978ppm; The expansion coefficient α and The thermal conductivity λ obtain the minimum values αmin=86.283°C⁻¹ and λmin=0.028W/(m·K), respectively; The acoustic loss and the sound absorption coefficient achieved maximum values of 9.1 dB and 0.88 at a frequency of 2000 Hz, respectively. Under the conditions of this paper, the optimal Na2O and K2O distribution ratio of Si-Ca-Mg-Al-Fe-B multi-component fiber glass system cotton was 16wt% and 0.5wt%, further demonstrating the feasibility of producing high water resistant microfiber glass by the centrifugation with synchronous water-repellent atomization spray in practical production applications.

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