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Infrared cooling properties of cordierite*

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Cordierite ($Mg_2Al_4Si_5O_{18}$) is known for its good thermal shock resistance and it is widely used to improve thermal shock properties of materials. We found that cordierite has good infrared heat dissipation performance. This performance provides an additional means of heat dissipation to assist in the cooling of the metal surface. Spectroscopic tests show that cordierite reflects sunlight in the visible range and emits infrared in the far infrared range, making it potential candidate as an infrared radiative cooling material for daytime use.

Keywords: daytime radiative cooling, cordierite, infrared emitting

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1. Introduction

Radiative cooling is increasingly attracting attention. This heat dissipation method takes advantage of sinking exhaust heat remotely, especially the heat sink of outer space, which has a temperature of 3 K. This method requires no further energy, which meets the requirements of saving energy.[1–3] Radiant cooling provides cooling power of up to 100 W/m^2 under suitable atmospheric conditions[4,5] thanks to the radiation wavelength peak of the object at ambient temperature (about 300 K) at the atmospheric radiation window. Although earlier studies have yielded good results, they have focused on infrared heat-emitting materials used at night,[6–14] due to the fact that the materials with high infrared radiation in the atmospheric window do not achieve intense solar reflection during the day.[15,16] In recent years, with the advent of some infrared metamaterials, this problem has been solved. Consequently, the research of daytime radiative coolers has now become the focus of attention.[17–19] For example, Zhai *et al.* fabricated a membrane, which dissipates heat by metamaterials combining polymethylpentene and quartz spheres. This membrane can reflect most energy of the sunlight and keeps the infrared emission in atmosphere window (8 μm$\sim$ 13 μm). The membrane can then cool down the object in daytime.[20] Goldstein *et al.* designed a sky radiation daytime cooler, which reduced the electricity consumption of a building up to 21%.[21,22] However, these metastructures rely on polymers, which are rather vulnerable under the penetration of ultraviolet rays in the sun's spectrum.

Here, we show that cordierite performs well as an infrared radiative cooler. It may also be a potential daytime radiative cooler, which has a low absorption in the visible light band and a good emissivity in the infrared band from 8 μm $\sim$ 13 μm.[23] Considering that cordierite also has good thermal shock resistance,[24,25] it may also be used for infrared radiating in high temperature environments, such as re-entry orbiters and high-speed aircraft, to help their cooling.

2. Experimental

Cordierite samples were prepared using a sol–gel method.[26,27] Tetraethyl silicate (TEOS) was used as the silicon source, and nitrates were used as the Mg source and the Al source. First, TEOS was dissolved in alcohol ethanol by volume ratio of 1:2, and then a magnetic stirrer was used to fully mix the two. The corresponding stoichiometric ratio $Mg(NO_3)_2{\cdot}6H_2O$ and $Al(NO_3)_3{\cdot}9H_2O$ were dissolved in water and stirred well. The magnesium nitrate solution was then added dropwise to the TEOS alcohol solution while keeping the solution stirred. The aluminum nitrate solution was added after magnesium nitrate solution. Nitric acid was added to the solution to adjust the pH to 2.5. The mixture was stirred for 3 hours and then placed in a ceramic container at 500 °C to fully oxidize the organic matter. The ashes were then thoroughly grounded and placed in a corundum ark and sintered at 1250 °C.

The composition of the sample was characterized by x-ray diffraction (XRD, Rigaku D / max-2400X) with a test angle (2θ) from 5° to 60°. The microstructure of the sample was characterized by scanning electron microscopy (SEM, Hitachi TM-1000). The ultraviolet-visible (UV-Vis) reflection absorption spectra of the samples were recorded on a Hitachi

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UV-4100 UV/Vis/NIR spectrometer. Infrared transmission absorption spectrum was recorded by a Fourier transformer infrared (FTIR) spectrometer (Thermo Scientific Nicolet iN10). The heat dissipation performance of the sample was tested as follows. Cordierite powder was first coated on a metal sheet (Fe or Cu), and the coated and uncoated metal sheets were heated to 200 °C and then left to cool naturally by air. The temperature of the metal sheets (at the bare metal side) was continuously recorded with an NR-82533B thermocouple until it drops to 50 °C. The time it takes to reach the specified temperature was recorded. The metal sheets were of the same size (8 cm×4 cm×1 mm). To measure the cordierite thickness, cordierite was spread evenly over the steel sheet, and then the cordierite was pressed with another steel sheet to obtain a flat and uniform cordierite coverage. After measuring the whole thickness and the thickness of the steel sheets, the thickness of cordierite can be obtained.

3. Results and discussion

Figure 1(a) shows the XRD pattern of cordierite at room temperature. The sample was found to be a single-phase compound with no other impurity peaks observed. The detected XRD patterns can be calibrated by orthorhombic unit cells with space group Cccm, $a = 9.721$ Å, $b = 17.062$ Å, $c = 9.339$ Å. There is a lot of space in the cell of cordierite, which leads to its smaller deformation at different temperatures and therefore provides it with good thermal shock resistance. Figure 1(b) shows an SEM photograph of cordierite. The photograph shows that the sample was composed of particles of about 200 nm sintered. It is worth mentioning here that there were a large number of micron-scale micro-pores on the sample, which may be caused by oxidation or decomposition of raw materials.

Before discussing the system's cooling details, we will review the three common modes of heat transfer: heat conduction, convection, and thermal radiation. They work together or competitively determining the temperature of the object. We can write the total energy obtained by an object as follows:

$$Q = Q_{\text{cdin}} - Q_{\text{cdout}} + Q_{\text{cvin}} - Q_{\text{cvout}} + Q_{\text{a}} - Q_{\text{r}}, \tag{1}$$

where Q_{cdin} represents heat conduction induced energy gain, Q_{cdout} represents heat conduction induced energy loss, Q_{cvin} represents heat convection induced energy gain, Q_{cvout} represents heat convection induced energy loss, Q_{r} represents the heat radiation emission, and Q_{a} represents the heat radiation absorption. While the temperature increase ΔT of the final object is given by the following equation

$$\Delta T = Q/C, \tag{2}$$

where C is the heat capacity of the object. Then we discuss the characteristics of different contributions. The overall energy change can be obtained from the integral of the heat flow ($Q = \int q(t)\,\mathrm{d}t$, q is the heat flow and t is the time). Thermal conductivity is described by Fourier's law[28,29]

$$q = -\kappa A\,\mathrm{d}T/\mathrm{d}x, \tag{3}$$

where q is the heat flux, κ is the thermal conductivity, A is the effective cross-sectional area for heat conduction, and $\mathrm{d}T/\mathrm{d}x$ is the temperature gradient in the direction of heat flow. The heat convection capacity is

$$q = mAC_{\text{p}}(T_2 - T_1), \tag{4}$$

where q is the heat flux, m is the fluid mass per unit area passing through, A is the convection cross-sectional area, C_{p} is the specific heat capacity of the fluid, and T_1 and T_2 are the temperatures at the outflow and inflow. For the sake of reunification, the thermal radiation capability can be written as[30]

$$q = \varepsilon\sigma AT^4, \tag{5}$$

where ε is the relative (blackbody) emissivity, σ is the Stefan constant, A is the area of radiation, and T is the absolute temperature. It is actually obtained by multiplying the Stefan–Boltzmann's Law by area. Based on Eqs. (3), (4), and (5)

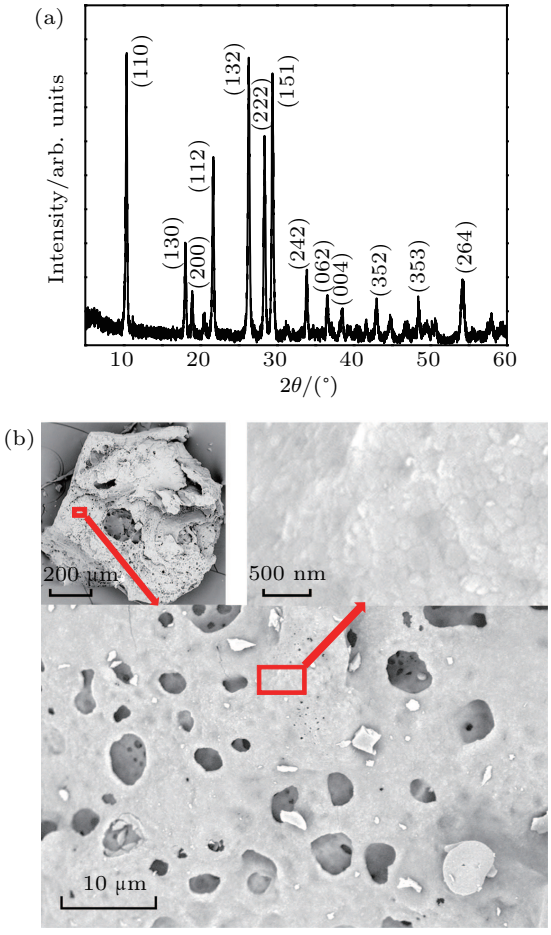


Fig. 1. (a) XRD pattern of cordierite, black curve for the test data, blue lines for the PDF card peak position. (b) SEM picture of cordierite.

we can compare the characteristics of different heat transfer modes. Considering all contributions vary with area, we can discuss the three behaviors across a particular area. The first is heat conduction—it can be seen that the weight become higher when the temperature difference between the two ends of the conduction region or the conductivity becomes higher. The second is convection—it can be seen that the weight become higher when the temperature difference between the two ends of the convection zone or the transferred heat capacity of the fluid becomes higher. However, the third, heat radiation, is rather different. The weight becomes higher when the sample is hotter or more emissive. According to the Kirchhoff's law of thermal radiation,[31,32] the relative emissivity coefficient and the electromagnetic wave absorption rate of sample at thermal equilibrium are same. In other words, the higher the infrared emissivity, the higher the infrared absorption.

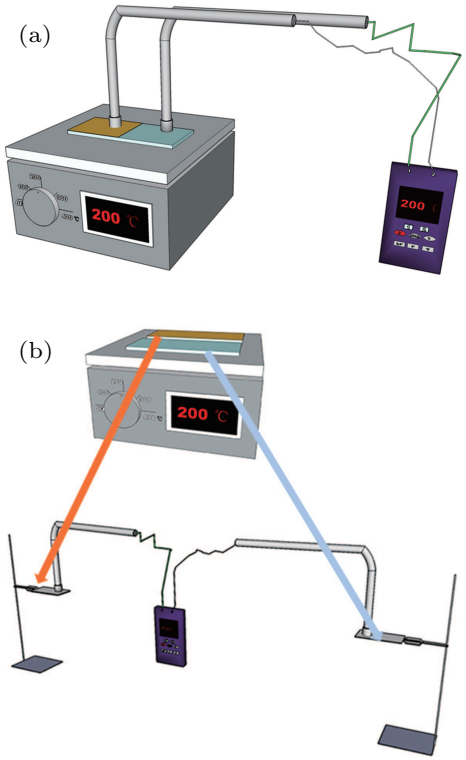


Fig. 2. Schematic illustration of the two sets for the cooling effect of cordierite. The blue sheets represent the metal sheets and the orange sheet represents the cordierite covered on the metal sheet. (a) Set A: Put the piece of copper sheet onto the hot plate and cover half of the copper sheet with cordierite. When the metal sheet reaches thermal equilibrium, measure the temperature of the cordierite and bare metal. (b) Set B: Put two pieces of metal sheets onto the hot plate and cover one piece of the metal sheet with cordierite. After reaching thermal equilibrium, two sheets were moved to the iron supports and cooled by air. The temperature of the two sheets and corresponding time were recorded then. The illustration in the upper right corner shows the difference between the infrared radiance of two different metal plates.

In this experiment, the hot metal sheets dissipate the heat by the air (heat conduction and convection) or by its own infrared radiation. Figure 2 shows the configurations of the cooling system we used. In Set A as shown in Fig. 2(a), we applied cordierite to the surface of the metal sheet and place the metal sheet on a hot plate and temperature of the hot plate was kept at 200 °C. The sheet reached thermal equilibrium after 1 hour. We were able to determine the temperature of the exposed cordierite and the exposed metal. We used this method to test the drop of cordierite temperature, and the cordierite was cooler than the exposed metal, as shown in Fig. 3(a). There are probably two possible reasons for this decline. One may be due to the cooling effect caused by infrared radiation, and the other possibility may be due to the poor thermal conductivity of cordierite. So we designed Set B, as shown in Fig. 2(b). This time we used two sheets of metal and covered one with cordierite. The two sheets were placed on a hot plate and moved to iron supports after reaching thermal equilibrium on the hot plate (200 °C). The thermal conductive heat dissipation of the support points cancels each other and both the systems of control group and experimental group were exposed to the air. The heat dissipation effects of the bottom sides of the two systems offset each other. The overall heat dissipation effects of the top sides are combinations of the air dissipation and infrared radiation dissipation. Considering that the thermal conductivity of cordierite is lower than the thermal conductivity of metal, the cordierite covered metal sheet will dissipate less heat to the air by thermal conduction and heat convection. As shown in Fig. 3(b) and Fig. 3(c), we can see that the temperature reduction of metal sheets covered by cordierite has been significantly accelerated, which shows that the infrared radiation takes a considerable amount of heat. This result is easy to understand. We know that the infrared emissivity of smooth metal surfaces is low (emissivity about 0.1). So the heat is mainly transmitted to the air by means of heat conduction and then dissipated by the air by heat convection to the surrounding air. The infrared radiation heat dissipation contribution is low in this case. Cordierite (emissivity $0.8 \sim 0.9$[26,33]) provides additional heat dissipation of infrared radiation to metal surfaces, which helps cooling of hot metals.

Figure 3(d) shows the enhancement effect of cordierite on the heat dissipation of iron and copper sheets. As can be seen, the time it takes for the steel sheet to cool down from 200 °C to 50 °C is a bit longer than the copper sheet. The infrared emissivity of the steel sheet is the same with the copper sheet (emissivities are both 0.10 for polished unoxidized steel and copper[34]). It is worth noting that the state of the air around the metal sheets is basically the same. However, heat capacities of the two metal sheets are not the same ($3.756\ \mathrm{J\cdot cm^{-3}\cdot K^{-1}}$ for steel and $3.45\ \mathrm{J\cdot cm^{-3}\cdot K^{-1}}$ for the copper[35]), the steel sheet cooled more slowly than the copper sheet because there are more heat to be dissipated in the steel sheet. The infrared radiating ability gained by introducing cordierite significantly enhanced the heat dissipation capacity of the metal sheets. Though the introduction of cordierite showed better effect on steel sheet than on copper sheet, the ratios of the reduced time in relative to the bare metal sheets are actually the same. This shows that this cooling method maybe universal.

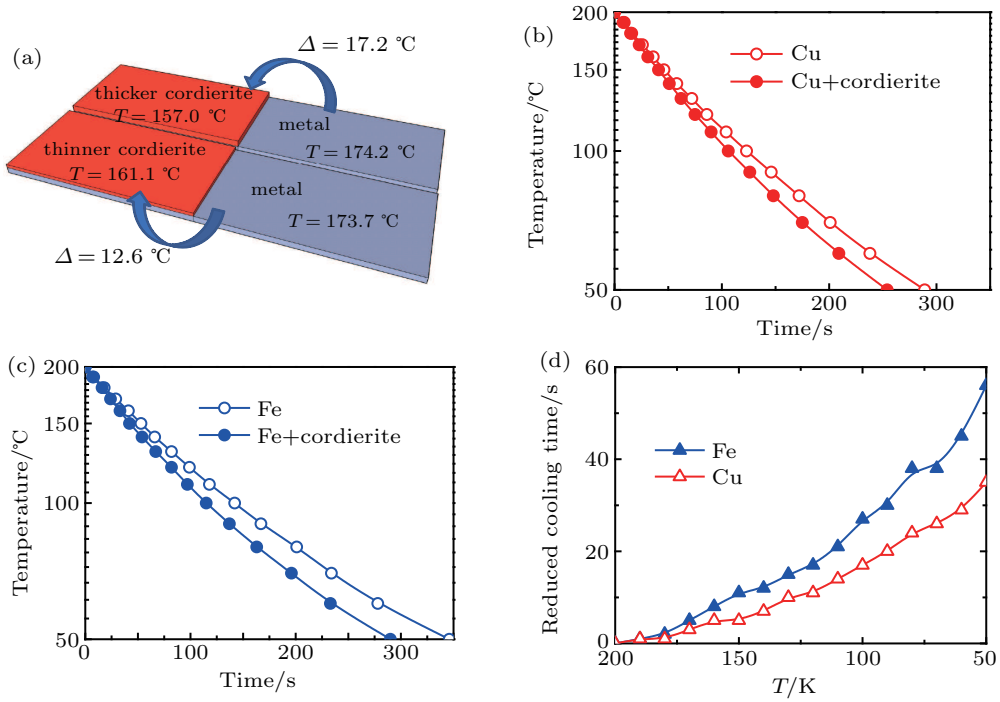


Fig. 3. (a) Final temperature of the cordierite and bare copper using Set A. Results of copper sheets covered by thicker cordierite and thinner cordierite are shown. (b) Comparison chart of temperature reduction effect of cordierite-covered copper sheet and pure copper sheet using Set B. (c) Comparison chart of temperature reduction effect of cordierite-covered steel sheet and pure steel sheet using Set B. (d) Reduced cooling time by covering cordierite of panels (b) and (c).

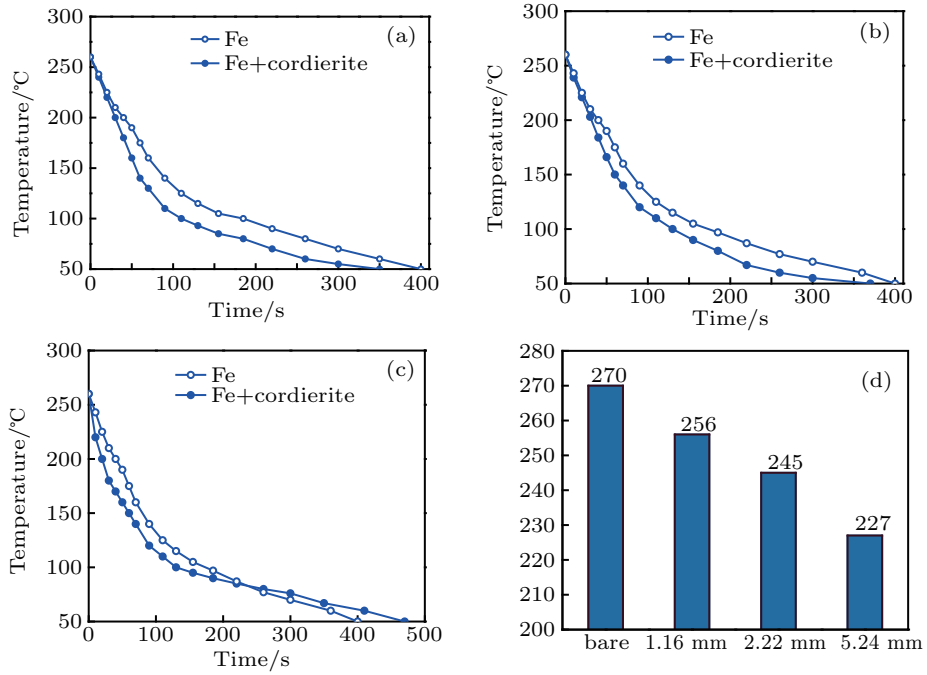


Fig. 4. (a) Comparison chart of temperature reduction effect of 1.16-mm cordierite-covered steel sheet and pure steel sheet using Set B. (b) Comparison chart of temperature reduction effect of 2.22-mm cordierite-covered steel sheet and pure steel sheet using Set B. (c) Comparison chart of temperature reduction effect of 5.24-mm cordierite-covered steel sheet and pure steel sheet using Set B. (d) Maximum heating temperature of bare steel sheet and cordierite-covered steel sheet of different thicknesses.

In the following experiments, we tested the infrared cooling performance of cordierite at high temperature. First, we heated the steel sheets on the heating stage to 260 °C, and the steel sheets were covered with cordierite equably in one side of the sheet in advance and the changes on both sides of the steel sheet were recorded. Through cooling, we obtained the cordierite cooling curve at high temperature. As shown in Fig. 4(a), at higher temperature, cordierite steel sheet covered by cordierite cooled to 50 °C faster than bare steel sheet. Therefore, it is proved that cordierite has excellent infrared cooling property at higher temperature (> 200 °C). Considering the influence of cordierite thickness on the cooling prop-

erty, we designed the cooling experiment of cordierite with different thickness on steel sheets of the same geometric size. We covered cordierite of different thicknesses at 1.16 mm, 2.22 mm, and 5.24 mm respectively. After heating the three steel sheets to the same temperature, the three steel sheets were cooled recording the data, as shown in Fig. 4(a), Fig. 4(b) and Fig. 4(c). It can be clearly observed that the thicker cordierite is, the worse its cooling performance is. When it reaches a certain thickness, its cooling performance is inferior than the bare steel sheet. This happens because the effective area of radiation of cordierite does not change with the increase of cordierite thickness, so the heat dissipation ability of infrared radiation to metal surfaces did not change. However, the cordierite is a porous structure, which shows low thermal conductivity. So when the cordierite is thick enough, thermal radiation will be masked by the poor thermal conduct, resulting in a thermal insulating effect at lower temperature. However, at higher temperature, the thermal radiation will be extremely accelerated, which will override the thermal insulating effect. This understanding can be proven by thermal imaging comparison chart. The dotted lines in the Fig. 5 are three steel sheet with the same size and the same mass. The upper part of the sheet is covered with cordierite of different thicknesses. The thickness from left to right is 1.16 mm, 2.22 mm, and 5.24 mm. It can be clearly seen from the Fig. 5 that the surface radiation of cordierite decreases with the increase of its thickness. Considering that the infrared emissivity did not change, the one with thicker cordierite surely has a cooler surface, which can be attributed to the thermal insulating effect, and the insulation will be enhanced with thicker cordierite.

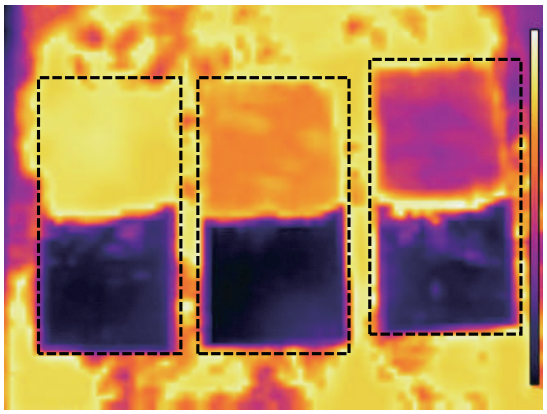


Fig. 5. Thermal imaging comparison chart.

To show the infrared cooling performance of cordierite more specifically, we calculate its average heat dissipation power by taking Fig. 4(a) as an example. The specific heat capacity of the steel sheet we used is 0.46×10^3 J/(kg·°C). The mass of the steel sheet is 38 g. When steel sheet cooled from 250 °C to 50 °C, it gives off heat to 3496 J. The difference in the time that it takes between cordierite-covered steel sheet and the bare steel sheet to cool to the same temperature is 50 seconds. Therefore, the average heat dissipation power of cordierite is calculated to be 70 W. The traditional computer CPU power is 50 W ∼ 100 W, so this enhanced cooling effect can meet the requirements of the CPU. So this is a powerful tool for the cooling of electronics.

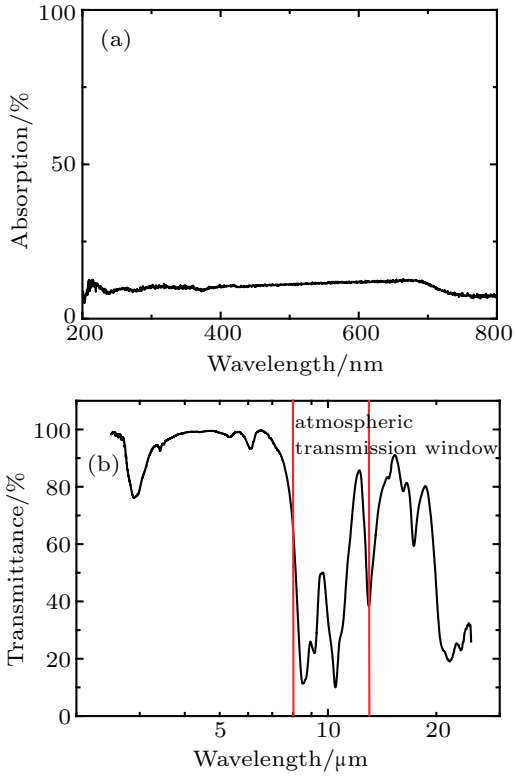


Fig. 6. (a) UV-Vis-IR reflection absorption spectra of cordierite. (b) FTIR transmission spectra of cordierite.

Recently, daytime radiative cooling has become a very popular topic.[20–22,36,37] Infrared cooling during the day requires the system to reflect most of the sunlight and to maintain sufficient infrared emissivity in the atmosphere window. This happens because, according to the Rayleigh formula, the temperature of the sun is about 6000 °C, so the thermal radiation is mainly concentrated in the visible region, and the energy in the atmospheric window band is very low. In stark contrast, at room temperature (300 K), the object's thermal radiation is concentrated in the atmospheric window area. Therefore, reflecting the solar radiation heat away, while maintaining the high emissivity in the infrared band, materials can be cooled during the daytime. Figure 6(a) shows the reflection absorption spectrum of cordierite. It can be seen that its absorption is extremely low in the visible wavelength range, and the main energy is reflected. Figure 6(b) shows the infrared transmission absorption spectrum of cordierite. Tests show that there is a clear infrared absorption of cordierite at the atmosphere window. According to Kirchhoff's law, the emissivity is equal to the absorption, so infrared tests show that there is an effective infrared emission of cordierite at the atmospheric window.[31,32] Combined with visible light reflection absorption spectra and infrared transmission absorption

spectrum analysis, we believe that cordierite is a good choice for daytime radiative cooling materials. It is worth mentioning that the existing daytime radiative cooling systems are mostly complex nanosystems, which hindered their applications. Meanwhile, a lot of daytime radiative cooling materials utilize the transparent infrared polymer materials, and the polymer materials have a serious aging problem under sunlight. There is no shortage of cordierite in this regard. It is an inorganic material that can withstand high temperatures, UV, *etc*., and thus can be more widely used in outdoor environments. Cordierite has also been used as an additive to enhance the thermal stability of the material due to its good thermal shock resistance.[24,25] Therefore, in the future, cordierite, as an infrared heat-radiating material is very likely to be applied to harsh environments.

4. Conclusions

In summary, we investigated the microstructure, heat dissipation and spectral absorption properties of cordierite. The results show that cordierite has good infrared radiating performance and can accelerate the cooling ability of the metal sheet. It also has good reflectivity in the visible band and good emission in the far infrared band. Consequently, it may be a good daytime radiative cooling material.

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