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Research Progress of Radiation Refrigeration Materials

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Abstract. Radiation cooling radiates heat to outer space through the atmospheric window. There is no energy consumption for the effect of refrigeration. Materials are the key to realizing radiation refrigeration. This paper mainly focuses on radiation refrigeration materials, summarizing some types of requisite materials, including the progress of making radiation refrigeration materials and the kinds of products. Furthermore, it also analyses some radiation effects that various radiation refrigeration products will bring, such as radiation refrigeration film and radiation refrigeration coating. Finally, it looks forward to the future development trend of radiation refrigeration materials.

Keywords. Radiation refrigeration, radiation refrigeration materials, radiation refrigeration film, radiation refrigeration coating

1. Introduction

Radiation refrigeration is an environmentally friendly and passive refrigeration method, which radiates the heat in the object through the transparent window of the atmosphere in the form of electromagnetic waves, in order to gain the effect of refrigeration without consuming other forms of energy. Radiation refrigeration requires special environmental conditions. Firstly, the tropospheric temperature is much lower than the annual ambient temperature in most parts of the earth. The troposphere acts as natural cold storage. Secondly, the atmosphere on earth is a natural transparent window in the 8-13μm band at ambient temperature, which can connect the thermal radiation of objects with the low-temperature troposphere [1, 2]. Simultaneously, with the aim of achieving the cooling effect, it is a necessity to radiate heat with high permeability or emit heat with high emission to reflect sunlight in the atmospheric window band. Therefore, to realize radiation refrigeration, materials need to reflect sunlight radiation and emit high emissions in the 8-13μm band [3]. Figure 1 shows the thermal radiation process of earth's surface objects. Radiation refrigeration materials are the key to realizing object radiation refrigeration. This paper summarizes the types of radiation refrigeration materials, the research progress of various radiation refrigeration materials, the types and application fields of radiation refrigeration products, and looks forward to the future development trend of radiation refrigeration materials.

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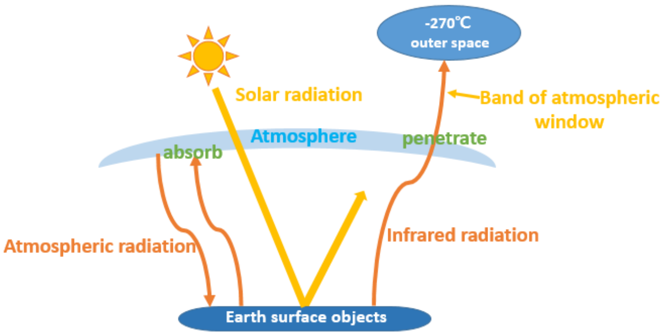

Figure 1. Schematic diagram of thermal radiation from earth surface objects.

2. Types of Radiation Refrigeration Materials

2.1. Classification by Material

Radiation refrigeration materials include monomer materials, composite materials, nanophotonic materials， and porous materials [4]. Monomer radiation refrigeration materials include polymer radiation refrigeration materials [5], inorganic radiation refrigeration materials [6], silicide radiation refrigeration materials [7], and gaseous radiation refrigeration materials [8]. Since the monomer radiation refrigeration materials are limited to the application of radiation scenes at night, the composite material composed of two or more materials is studied. The structure of the composite material is modified to have the function of radiation refrigeration in the daytime [9]. Nanophotonic radiation refrigeration materials include single, double, three， or more layers [10]. No matter which nanophotonic material can reflect sunlight and high emission in infrared window band, its application is limited due to its high manufacturing cost. Porous radiation refrigeration materials include nanoporous polyethylene, porous polymer coating, and porous alumina [11]. Nanoporous materials have high visible light reflectivity and infrared emissivity. Porous polymer coatings have high light reflectivity and thermal emissivity and can be sprayed on various object surfaces to achieve an efficient radiation refrigeration effect in the daytime. Porous alumina has excellent emissivity, porous alumina film has efficient radiation refrigeration characteristics and low manufacturing cost, so it is suitable for large-scale production. Figure 2 shows the classification of radiation refrigeration materials.

2.2. Classification by Function

Radiation refrigeration materials include daytime radiation refrigeration materials and night radiation refrigeration materials according to their functions. The research of radiation refrigeration materials began in the 1950s. Early studies focused on radiation cooling at night. In the 1970s, researchers began studying daytime radiation materials [12]. Daytime radiation refrigeration uses the cold space as the cold source. By adjusting the optical and infrared response, the material has a very high reflectivity (> 90%) in the sunlight band (0.3μm-2.5μm) and reduces the heat absorbed by the object from the sunlight. At the same time, it promotes the material to have high emissivity (> 90%) in the 8 μm-13μm band of atmospheric infrared transmission window and radiant heat into the universe through the atmosphere, to realize the refrigeration effect that the surface temperature of the object is lower than the ambient temperature under direct sunlight. The early preparation of the radiation refrigerator mainly used polymer film, white coating, oxide film, and other materials. These materials have low emissivity in the 8-13μm band, and the radiator has insufficient reflection ability to solar radiation. In recent years, researchers have been deepening the research on radiation materials, using layered structures, nanoparticles or pores to enhance the selective radiation ability of materials in the 8-13μm band [13].

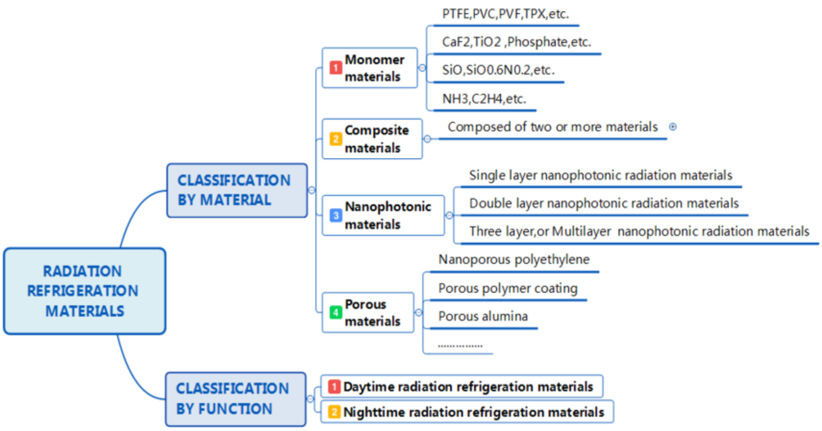

Figure 2. Classification of radiation refrigeration materials.

3. Research Progress of Radiation Refrigeration Materials

3.1. Research progress of Monomer Radiation Refrigeration Materials

Researchers have found a variety of monomer radiation refrigeration materials with a high absorption rate in the 8-13μm band, such as PTFE, CaF_2, silicide, phosphate, and so on. Yang et al. studied the radiation refrigeration effect of PTFE and CaF_2. The temperature difference between PTFE refrigeration space and environment reaches 8.1-11.5℃. The maximum temperature difference between CaF_2 refrigeration space and environment reaches 8-11℃ [14]. Granqvist et al. studied the radiation refrigeration effect of silicide radiation refrigeration materials and found that SiO has strong lattice absorption near 10μm. In the night test, the SiO cooler cooled to 13.8℃ lower than the ambient temperature, while the blackbody cooler also cooled to 13.4℃ lower than the ambient temperature. Compared with the blackbody cooler, the emission intensity of SiO in the atmospheric window is not strong enough [15]. Granqvist et al. also studied the silicon nitride cooler coated with aluminum on the back. A layer of $SiO_{0.6}N_{0.2}$ film with a thickness of 1.34 μm is attached to the aluminum back reflection film, which can achieve low reflectivity in the atmospheric window. If the reflectivity of the silicon nitride cooler surface is lower, the absorption rate of the cooler's surface is higher. In the test at night, the silicon nitride cooler cooled to 16℃ lower than the ambient temperature and 2℃ lower than the blackbody cooler [16]. Xu et al. studied the mixed-phase crystal material of magnesium phosphite, solar radiation reflectance is as high as 94.60% in the

200-2500 nm solar spectral region, emissivity is as high as 0.93 in the 1-22 mm infrared spectral region. Magnesium phosphate crystal has excellent reflectivity and emissivity. It is an ideal functional filler for radiation refrigeration coating. $Mg_{11}(HPO_3)_8(OH)_6$ crystal is used as the functional filler to prepare the radiation refrigeration coating. Under daytime sunlight radiation, the air temperature under $Mg_{11}(HPO_3)_8(OH)_6$ crystal coating is 2-4°C lower than that under TiO_2 and $CaCO_3$ coating of the same specification, and 6-8°C lower than that under high reflectivity aluminum foil. Under ideal conditions, the radiant cooling power of $Mg_{11}(HPO_3)_8(OH)_6$ crystal coating can reach 45.36 W/m^2 [17].

Some gases can also be used as radiation coolers. An ammonia gas plate will cool to below the ambient temperature at night [18]. Ethylene also shows high emissivity in the atmospheric window. When encapsulating in an infrared transparent container, C_2H_4 is a good radiation cooler, which can reach 10°C lower than the ambient temperature in the daytime without direct sunlight [19].

Table 1 compares the refrigeration characteristics of monomer materials. Monomer materials have good radiation refrigeration characteristics at night, earth surface objects can be cooled to 16°C below the ambient temperature.

Table 1. Refrigeration characteristics and effects of monomer materials.

Monomer material	Refrigeration characteristics	Refrigeration effect
PTFE	Normal Emittance is 0.9282	8.1-11.5°C lower than the ambient temperature
CaF_2	Normal Emittance is 0.9279	8-11°C lower than the ambient temperature
SiO	Strong lattice absorption near 10μm	13.8°Clower than the ambient temperature in the night test
$SiO_{0.6}N_{0.2}$	Aluminum plating on the back can achieve low reflectivity in the atmospheric window	16°C lower than the ambient temperature in the night test
$Mg_{11}(HPO_3)_8(OH)_6$	Both reflectivity and emissivity are excellent	2-4°C lower than TiO_2 and $CaCO_3$ coating, and 6-8°C lower than high reflectivity aluminum foil
NH_3	Ammonia gas plate has the characteristics of radiation refrigeration at night	Cool to below the ambient temperature at night
C_2H_4	When encapsulated in infrared transparent container, it has good radiation refrigeration performance	10°C lower than the ambient temperature in the daytime without direct sunlight

3.2. Research Progress of Composite Radiation Refrigeration Materials

Because the monomer cannot be used for daytime radiation refrigeration, researchers studied the composite radiation refrigeration material composed of two or more materials, modified its structure to make it have radiation refrigeration performance in the daytime.

In 2017, Zhao et al. randomly embedded resonant polar dielectric microspheres in the polymer matrix, the metamaterial is 100% transparent in the solar spectrum, and has an infrared emissivity greater than 0.93 in the atmospheric window. A layer of silver is coated on the backing surface of the metamaterial, which has a maximum radiation cooling power of 93 W/m^2 in direct sunlight [20].

In 2019, Zhang et al. prepared a series of polymer films containing inorganic additives by electrospinning and coating, studied the radiation cooling effect of different polymers or different additive films. The results show that the polymer films with ZnO, CaF_2, and ZnS have an excellent refrigeration effect. The polymer film added CaF_2

prepared by the coating method has the best radiation refrigeration effect. When 0.2g CaF_2 (polyethylene mass is 5.0 g) was added to the polyethylene film, the measured equilibrium temperature dropped from 37.1℃ to 34.0℃. When 0.0234g CaF_2 (3.6g PVC mass) was added to the PVC fiber film, the measured equilibrium temperature dropped from 37.8℃ to 35.2℃ [21].

In 2020, Zhang et al. invented a daytime radiation cold water coating suitable for building surfaces, adding fluorescent materials to titanium dioxide nanoparticles, reducing the absorption of sunlight by "multiple scattering + fluorescence emission", the effective reflectivity increased from 89.8% to 93.4%. At the same time, by adding glass microspheres with wide particle size into the coating, phonon enhanced resonance realizes ultra wideband infrared radiation. The emissivity of 3-50μm is 90%, the emissivity of the infrared atmospheric window band is 95%-96%. Wide-spectrum radiator emits heat to outer space through the atmospheric window continuously, fully exchanging dynamic heat with atmospheric longwave radiation. Through triple synergy, the newly developed SDRC coating can not only realize refrigeration, but also reduce the temperature difference between day and night on the building surface and inside, using the aluminum plate as the coating substrate, the coating surface is 6±1℃ lower than the ambient temperature at noon, the coating surface is 4±0.3℃ lower than the ambient temperature at night [22].

Table 2. Refrigeration characteristics and effects of composite materials.

Composite material	Refrigeration characteristics	Refrigeration effect
Randomly embeds resonant polar dielectric microspheres in a polymer matrix	Infrared emissivity greater than 0.93	Radiant cooling power can reach 93 W/m^2 in direct sunlight
Polymer film containing inorganic additives	The polymer films with ZnO, CaF_2,and ZnS have an obvious radiation refrigeration effect	Adding inorganic additives can lower 3℃ again
Fluorescent materials and wide particle size glass microspheres were added to titanium dioxide nanoparticles	The effective reflectivity increased from 89.8% to 93.4%,90% emissivity in 3-50 micron band, 95%-96% emissivity in the infrared atmospheric window	6±1℃ lower than the ambient temperature at noon, 4±0.3℃ lower than the ambient temperature at night.
polymer nanofiber (ES PEO) film	The chemical bond vibration peak of polyethylene oxide (PEO) overlaps with the main band channel of thermal radiation	5℃ lower than the ambient temperature under sunlight, 7℃ lower than the ambient temperature at night

In 2020, researchers from Nanjing University studied “polymer nanofiber (ES PEO) film”, the chemical bond vibration peak of polyethylene oxide (PEO) overlaps with the main band channel of thermal radiation, so that heat can only go out. ES PEO film can achieve the refrigeration effect of 5℃ lower than the ambient temperature under sunlight, and about 7℃ lower than the ambient temperature at night [23].

Table 2 compares the refrigeration characteristics of composite materials. Composite material has a good refrigeration effect in the daytime, earth surface objects can be cooled to 7℃ below the ambient temperature.

3.3. Research Progress of Nanophotonic Radiation Refrigeration Materials

In 2014, Fan et al. Deposited multilayer nanophotonic crystals on the silver substrate, 5℃ lower than the ambient temperature in the daytime without consuming electric energy [24].

In 2017, Zhai et al randomly embedded SiO_2 microspheres (about 8μm in diameter) into polymethylpentene, made into a 50 μm thick film, which radiates energy outward in the form of infrared electromagnetic waves to achieve the effect of refrigeration, the emission wavelength range is 8-13 μm, and the infrared emissivity is as high as 0.93, closing to the ideal blackbody. A 200 nm thick silver film coated on the back of the film, can obtain up to 96% solar reflectivity and further improve the refrigeration effect of the film material. It has a radiation refrigeration power of up to 93 w/m^2 under direct midday sunlight, which can reduce the temperature of the object in contact with it by 10-16℃ [25].

In 2017, Zhi et al. Proposed an acrylic resin double-layer coating embedded with titanium dioxide and carbon black particles. The top layer is responsible for reflecting solar radiation, and the bottom layer emits heat in the atmospheric transparent window. Acrylic resin has high transparency in the spectral range and does not absorb additional energy such as sunlight. Rutile titanium dioxide does not absorb solar radiation in most solar spectra and has a high refractive index to reflect solar radiation. Carbon black particles have high emission and heat emission in the atmospheric window. Test titanium dioxide particles of different sizes on the top layer, that the particles with a radius of 0.2 μm could provide the best cooling performance. More than 90% of solar radiation can be reflected, and the average emissivity of the atmospheric transparency window exceeds 0.9 in most directions. The net cooling power during the day exceeds 100 W/m^2 at ambient temperature [26].

Table 3. Refrigeration characteristics and effects of nanophotonic materials.

Nanophotonic materials	Refrigeration characteristics	Refrigeration effect
Multilayer nanophotonic crystals on the silver substrate	Nanophotonic crystal structure makes it have good cooling performance	5℃ lower than the ambient temperature
SiO_2 microspheres (about 8 μm in diameter) into polymethylpentene	Up to 96% solar reflectivity	A radiation refrigeration power of up to 93 W/m^2
Acrylic resin double-layer coating embedded with titanium dioxide and carbon black particles	The average emissivity of the atmospheric transparency window exceeds 0.9 in most directions	The cooling power during the day exceeds 100 W/m^2 at ambient temperature
Optimized BN, SiC, and SiO_2 gratings	High emittance in the 8-13 μm atmospheric transparency window	The average radiation cooling power during the day is 55 W/m^2 under high radiation intensity

In 2018, Hervé et al. proposed a radiation scheme combining grating and multilayer thermal control capability. Optimized BN, SiC, and SiO_2 gratings have high emittance in the 8-13 μm atmospheric transparency window. Putting a metallic electrolyte under the grating can promote reflectivity close to 100% in the solar spectrum and enhance the emissivity in the atmospheric window band. Control the surface charge of polar materials and promote the polarization of surface phonons. The multilayer structure produces resonance radiation, which helps to radiate heat energy. The radiation cooling power of this structure is as high as 80 W/m^2 at night, and the average radiation cooling power during the day is 55 W/m^2 under high radiation intensity [27]. This method has a simple preparation process and advantages over other multilayer structures.

Table 3 compares the refrigeration characteristics of nanophotonic materials. Nanophotonic material has a good refrigeration effect in the daytime, earth surface objects can be cooled to 5℃ below the ambient temperature.

3.4. Research Progress of Porous Radiation Refrigeration Materials

Researchers have studied porous radiation refrigeration materials such as nanoporous polyethylene, porous polymer coating, and porous alumina. Nanoporous materials have high visible light reflectivity and infrared emissivity, porous polymer coating has high light reflectivity and thermal emissivity, and porous aluminum oxide has excellent emissivity.

Cui et al. studied nanoporous metalized polyethylene and prepared nanophotonic structure textiles through modification. The textile can be cooled passively. At 12 μm thick nanometer polyethylene film is plated with a layer of nanoporous Ag film. Then press the side of nanoporous Ag film onto the surface of the cotton fabric to obtain the composite film. The lowest infrared emissivity of nanoporous metalized polyethylene fabric is 10.1% on the nonmetallic polyethylene side. This fabric has good wear resistance as ordinary fabric. Compared with traditional cotton fabric, nanoporous metalized polyethylene fabric can reduce the ambient temperature by 7.1°C. Nanoporous metalized polyethylene fabric is much better than all existing radiation-heated fabrics. This wearable nanoporous metalized polyethylene fabric has an excellent local heating capacity. However, the use of Ag makes the manufacturing cost higher [28].

The Columbia University research team has studied a simple, scalable， and cheap phase conversion process to produce a layered porous polymer coating with excellent visible light reflectivity and heat incidence. The layered porous polymer coating can achieve a radiation cooling effect lower than the ambient temperature during the day. The visible reflectance of layered porous polyvinylidene fluoride-co- hexafluoropropylene is 0.96 ± 0.03, and the emissivity is 0.97 ± 0.02. At solar intensities of 890 and 750 W/m^2, the layered porous polymer coating can reach about 6°C below the room temperature, and the average cooling power is 96 W/m^2 [29].

Porous alumina is also a kind of radiation refrigeration material. Fu et al. used porous anodic alumina film in a daytime passive radiation refrigeration cooler. Alumina structure is optimized to have high absorptivity (emissivity) in the far-infrared atmospheric window. Alumina has strong phonon resonance absorption in the far-infrared window, but it is not directly suitable for cooling applications. Hole dilution helps to reduce the dielectric constant, to improve the impedance matching with the surrounding medium (air) [30]. Zhang et al. studied the porous alumina radiation refrigeration material.

The porous alumina film has excellent emissivity, which is 2.6°Clower than the ambient temperature. The cooling power density is 64 W/m^2 in the environment of direct sunlight (AM1.5) and 75% humidity. It also has almost the same cooling effect at night. The porous alumina film is suitable for mass-produced with low manufacturing costs [31].

Table 4 compares the refrigeration characteristics of porous materials. Porus material has a good refrigeration effect in the daytime, so earth surface objects can be cooled to 7.1°C below the ambient temperature.

Table 4. Refrigeration characteristics and effects of porous materials.

Porous materials	Refrigeration characteristics	Refrigeration effect
Nanoporous metalized polyethylene	The lowest infrared emissivity is 10.1% on the nonmetallic polyethylene side	7.1°C lower than the ambient temperature
Layered porous polyvinylidene fluoride co hexafluoropropylene	The visible reflectance is 0.96 ± 0.03, the emissivity is 0.97 ± 0.02	The average cooling power is 96 W/m^2, 6°C lower than the ambient temperature
The porous alumina film	The cooling power density is 64 w/m^2	2.6°C lower than the ambient temperature

At present, the radiation refrigeration film and radiation refrigeration coating are widely used, which have been used in building cooling, substation cooling, communication base station cooling, photovoltaic cooling (solar photovoltaic, concentrating photovoltaic, thermal photovoltaic), electronic product heat dissipation, and other fields [32, 33].

3.5. Radiation Refrigeration Film

Radiation refrigeration films include reflective refrigeration films and transmissive refrigeration films. Due to its reflection and transmission characteristics, the composite radiation refrigeration film has high reflectivity in the visible band of the solar spectrum and high emissivity in the atmospheric window band of 8-13 μm. The reflective radiation refrigeration film (FR1H100) of Radial-Cool advanced energy technologies Co. Ltd. can reduce the surface temperature to 5-10°C below the ambient temperature under midday sunlight and realize the evaluation refrigeration power of more than 100 W/m^2. It has applied to construction, industry, transportation, modern agriculture, power equipment, telecommunications, cold chain logistics, and other fields. Radiation refrigeration film has been used in the corridor bridge of the Xiaoshan airport terminal building. The energy-saving of a single corridor bridge is 55000 kWh/year, and the energy-saving rate of air conditioning is as high as 43.7%. Reflective radiation refrigeration film has been used in Singapore Changi Airport, and the annual power saving rate of the corridor bridge is about 40%. The reflective radiation refrigeration film has been used in Haneda airport, Japan. The temperature of the outer surface of the roof is 6.3°C, and the regional ambient temperature is 6.3°C.

3.6. Radiation Refrigeration Coating

Chen et al. selected the substation box for the application effect experiment of radiation refrigeration coating. The radiation refrigeration coating has an excellent cooling effect on the substation box and cabinet arranged outdoors. Under cloudy weather conditions, the temperature of the top outer surface of the test cabinet with radiation refrigeration coating is 11.4-18.6°C, the temperature of the inner surface of the top is 9.8-16.1°C and the air temperature in the cabinet is 4.1-5.7°C lower than that of the blank cabinet without any treatment. The application of radiation refrigeration materials improve the operation reliability, safety, and overload time of various equipment in the substation, reducing the time and frequency of air conditioning, and the consumption of air's power conditioning [34].

4. Conclusion and Prospect

To sum up, researchers have conducted in-depth research on the radiation refrigeration characteristics of various radiation refrigeration materials. In recent years, researchers have made many breakthroughs in composite materials, nanophotonic materials, and porous materials used in daytime radiation refrigeration. At the same time, according to the using scenario, radiation refrigeration materials have been made into many radiation refrigeration products. Radiation refrigeration films and coatings have been mature and applied to building cooling and heat dissipation of the communication base station.

For radiation refrigeration products, the control of material emissivity is not a complete decisive factor. Radiation refrigeration materials are only one of the factors to achieve efficient radiation refrigeration. The thermal insulation performance and weather conditions of radiation refrigeration devices also affect the performance of radiation refrigeration. The design of the radiation refrigeration device affects the non-radiation heat transfer coefficient. From the perspective of device design, the lower the non-radiation heat transfer is required, the better. Therefore, the radiation refrigeration system should be more insulated. Weather conditions will also affect the emissivity of the atmosphere. In sunny weather, the lower the water vapor content in the atmosphere will be better. It is easier to realize the effect of radiation refrigeration in dry areas. The radiation refrigeration effect is affected by materials, systems, and weather. In the future, researchers need to deeply study the influence law of various factors such as materials, systems, and weather conditions on the effect of radiation refrigeration through modeling and other means to promote the application of radiation refrigeration materials.

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