Research Article

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Research on materials of solar selective absorption coating based on the first principle

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Abstract: Based on the first principle, this paper studies the optical properties of Ni, Mo, CoO, and Cr_2O_3 according to the Materials Studio software. It is found that the absorptivity of Ni is low, while Ni has low emissivity. Hence, it can be used to reduce emissivity. The absorption rate of CoO is very high. Therefore, Ni and CoO are very suitable to be composed to make a solar selective absorption coating with high absorptivity and low emissivity. The mass ratio of Ni and CoO has a greater impact on the optical properties of the composite material, so the absorption–emission ratios of the composite material Ni–CoO at different mass ratios are calculated. The absorption–emission ratio is the highest when the mass ratio is 1:1, and the performance is the best, which is in good agreement with the result of the experiment. And we hope that our method will provide some help for the study of solar selective absorption composite coatings.

Keywords: solar selective absorption coating, the first-principle, optical performance, absorption–emission ratio

1 Introduction

As the global energy crisis and environmental degradation are becoming increasingly serious, solar energy has attracted widespread attention. However, solar energy

has a low light-to-heat conversion efficiency, so the utilization technology of solar energy should be developed. The solar selective absorption coating is the most core part of a photothermal energy system. Increasing the absorption rate and reducing the emissivity of the absorption coating is one of the most important ways to improve the efficiency of the solar system. Solar selective absorption coatings have been widely studied, and scholars at home and abroad have done a lot of work in this area. As early as the 1950s, Tabor developed a black nickel coating [1]. In 2011, Wang et al. optimized the black nickel coating [2]. At present, the more common coatings are multilayer gradient coatings [3], optical interference coatings [4], and ceramic composite coatings [5].

In recent years, metal-ceramic composite coatings have been widely studied due to the advantages of convenient performance improvement, low pollution, and low cost. Metal-ceramic films mainly use Al_2O_3 as a dielectric substrate. A large number of such coatings have been developed, such as Fe– Al_2O_3 [6], W– Al_2O_3 [7], Ag– Al_2O_3 [8], AlNi– Al_2O_3 [9], MoSi_2 – Al_2O_3 [10], AgAl– Al_2O_3 [11], Pt– Al_2O_3 [12], Ni– Al_2O_3 [13], Mo– Al_2O_3 [14], WTi– Al_2O_3 [15] and W–Ni– Al_2O_3 [16]. What's more, the film system of Al_2O_3 –WC/ Al_2O_3 –WC/ Al_2O_3 that was designed by Gao et al. [17] made the full ceramic coating possible.

In addition, there are cermet films that use other substances as dielectric substrates. Brashilia not only prepared the TiAlN/TiAlON/ Si_3N_4 coating [18] by DC reactive magnetron sputtering but also optimized the TiAlN/AlON coating [19] and the HfOx/Mo/HfO₂ coating [20]. Wang developed the first high-temperature spectral selective absorption coating of the Nb–NbN system [21]. Wang et al. developed the Mo– SiO_2 coating system [22]. The $\text{Ti}_{0.5}\text{Al}_{0.5}\text{N}$ –AlN coating was developed by Du et al. [23]. Rebouta designed the TiAlN/TiAlON/ SiO_2 composite coating [24] and also prepared the AlSiN/AlSiON dual-layer solar selective coating [25]. Cespedes et al. first studied a selective coating material based on Mo– Si_3N_4 [26]. There are also new AlCrNO coatings [27], Nb–TiO₂ absorbing coatings [28], AlMoN-based multilayer absorbing coatings [29], and new Mo/ZrSiN/ZrSiON/ SiO_2 solar selective absorbing coatings [30].

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Experiments are often expensive and time-consuming; so, numerical simulation research has become more extensive with the development of computer technology. Liu *et al.* [31] established a computer model to calculate the emissivity of the coating based on the Mie scattering theory and Kubelka-Munk model. Esposito S and Antonaia used a semi-empirical method to simulate and optimize the optical properties and high-temperature durability of Mo-SiO₂ [32] and W-Al₂O₃ [33] film systems by computer. Fang used Materials Studio software to calculate the reflectivities of materials [34]. Xin simulated the selective absorption coating with a hypothetical model of linear emissivity [35]. Sakurai *et al.* used the characteristic matrix method and genetic algorithm to optimize the multilayer structure and volume fraction of various metals of W-SiO₂ cermet [36]. Liu developed a new type of Mo/NiAlN/NiAlON/SiO₂ multilayer film solar selective absorption coating using Scout software [37]. Nuru *et al.* used a computer simulation program to calculate the appropriate complex refractive index of the Pt-Al₂O₃ bimetal ceramic structure [12]. Li established a metal-ceramic selective absorption coating emissivity model by programming with Labview software and carried out simulation experiments on the emissivity variation pattern of Mo-SiO₂ coatings to optimize the coating structure parameters [38].

At present, almost all selective absorption coatings are composite materials. To obtain a composite film with good optical performance, the selection and ratio of composite coating materials are very important. So, the optical properties of the materials are studied this time. Because most of these problems are studied through complex experiments, we want to replace experiments with simulations. The materials with good performance are selected for compounding and research ratio using the Materials Studio software. Finally, a composite material can be obtained which can be used to improve the optical properties of the selective absorption coating.

2 Calculation of the optical properties of Ni, Mo, CoO, and Cr₂O₃

The microstructure of a material determines its macroscopic properties. The electronic structure of a material is intrinsically related to many properties such as its optical properties and electrical conductivity. According to the first principle, some approximate treatments can be made from the perspective of microscopic nuclei and

electrons through quantum mechanics, which can predict the crystal structures, optical properties, density of states, and band structures of solids. This article mainly studies the electronic structure (band structure and density of states) and optical properties (reflectance, absorptivity, and emissivity) of materials, so the first principle is used as the theoretical basis.

For the common raw materials of cermet films, Ni, Mo, CoO, and Cr₂O₃ were selected for calculation and analysis. To select two materials with good properties from these four materials to prepare composite materials, the optical properties of each material need to be calculated. The CASTEP module of Materials Studio software is based on density functional theory (DFT) and can be used to study the surface chemistry, the bond structure, the density of states, the optical properties, and so on. It is suitable for a variety of lattice systems, including metal materials, ceramics, and semiconductors. This paper uses it to calculate the band structure and density of states of the unit cell. The first is to construct the crystal structure diagram of each substance through its space group, lattice constant, and atomic coordinates. Then, the most stable crystal structure of each substance can be obtained through optimizing. The lattice constants and space groups of Ni, Mo, CoO, and Cr₂O₃ cells are shown in Table 1. The crystal structures of the four substances are shown in Figure 1.

2.1 Band structure and density of states

The band structures and density of states of Ni, Mo, CoO, and Cr₂O₃ were calculated according to each crystal structure using the CASTEP module of Materials Studio, as shown in Figure 2.

It can be seen from Figure 2 that there are multiple energy bands in the band diagram of Ni that pass through the Fermi surface, indicating that it has good conductivity. Electrons of 3p orbital and 4s orbital of Ni can

Table 1: Lattice constant and space group of each substance

Substance	Lattice constant			Space group
	<i>a</i>	<i>b</i>	<i>c</i>	
Ni	3.5164	3.5164	3.5164	FM-3M (OH-5)
Mo	3.1656	3.1656	3.1656	IM-3M (OH-9)
CoO	4.2383	4.2383	4.2383	FM-3M (OH-5)
Cr ₂ O ₃	4.9364	4.93364	13.7984	R-3C (D3D-6)

Figure 1: Crystal structures of Ni, Mo, CoO, and Cr_2O_3 : (a) Ni, (b) Mo, (c) CoO, and (d) Cr_2O_3 .

transition to electrons of 3d orbital and absorb photons. But, the density of states of electrons in 3p and 4s orbitals is mainly concentrated below -1 eV , and the density of states in this range is smaller, so the absorption of photons greater than -1 eV will be small. So, the absorptivity of Ni won't be very large. Mo's 4p and 5s orbital electrons can absorb photons through transitioning to 4d orbital electrons. But Mo has very few 5s and 4p electrons, so the absorptivity won't be large.

2.2 Calculation of absorption emission ratio

We first calculated the reflectance using Materials Studio software, because the MS software couldn't directly calculate the absorptivity and emissivity of a substance. Then, we calculated the absorptivity and emissivity according to the corresponding formulas.

2.2.1 Reflection curve

For the crystal structures of Ni, Mo, CoO, and Cr_2O_3 , their reflectivities were calculated based on the density functional theory. As is shown in Figure 3, their reflection curves in the range of 0–5 eV are made, because the spectral band of solar energy is mainly in the range of 0.5–4.1 eV.

2.2.2 Calculation of absorption and emissivity

We used the formulas to calculate the absorptivities and emissivities of Ni, Mo, CoO, and Cr_2O_3 according to their reflectivities. The solar energy of 0.3–2.5 μm is selected to calculate the absorptivity to simplify the calculation process, because most of the solar energy is concentrated in the range of 0.3–2.5 μm , while that in the other bands is approximately 0. For objects with a temperature between room temperature and 1,000 K, the solar energy

Figure 2: Energy band structures and the density of states of Ni, Mo, CoO, and Cr_2O_3 . (a) Energy band structure of Ni, (b) the density of states of Ni, (c) energy band structure of Mo, (d) the density of states of Mo, (e) energy band structure of CoO, (f) the density of states of CoO, (g) energy band structure of Cr_2O_3 , and (h) the density of states of Cr_2O_3 .

Figure 3: The reflectivity curves of Ni, Mo, CoO, and Cr_2O_3 .

of $0.3\text{--}2.5\text{ }\mu\text{m}$ is selected to calculate the emissivity, because the wavelength of thermal radiation is mainly concentrated in this range. The temperature is uniformly selected to 800 K .

$$\alpha(T) = \frac{\int_{0.3}^{2.5} d\lambda S(\lambda)[1 - R(\lambda, T)]}{\int_{0.3}^{2.5} d\lambda S(\lambda)}, \quad (1)$$

$$\varepsilon(T) = \frac{\int_{2.5}^{25} d\lambda E(\lambda, T)[1 - R(\lambda, T)]}{\int_{2.5}^{25} d\lambda E(\lambda, T)}, \quad (2)$$

$$E(\lambda, T) = \frac{2\pi hc^2 \lambda^{-5}}{e^{hc/k_B \lambda T} - 1}, \quad (3)$$

where λ is the wavelength of the solar spectrum, $R(\lambda, T)$ is the reflectance, h is the Planck constant, c is the speed of light in a vacuum, and k_B is the Boltzmann constant. From Figure 4, $S(\lambda)$ is the density of the solar radiation energy flow (using the standard AM1.5). The absorptivities and emissivities of these four substances were calculated using the formulas of (1)–(3). And then their absorption–emission ratios could be calculated. The results are listed in Table 2.

However, in practice, when the absorptivity goes on increasing after a certain value, the emissivity will also increase significantly. It is difficult to measure the optical properties of a substance using one of the indicators because sometimes the increase in emissivity is even greater than the increase in absorptivity. Therefore, the absorption–emission ratio is used to estimate the performance of the coating. The larger the absorption–emission ratio is, the better the performance is. From Table 2, we can find the absorption–emission ratios: $\text{Ni} > \text{CoO} > \text{Mo} > \text{Cr}_2\text{O}_3$.

It is found that the absorption–emission ratio of Ni is the largest, but its absorptivity and emissivity are not

Figure 4: The density of Solar spectral energy flux (Standard AM1.5).

high, which indicates that the absorption performance of Ni is not good, but it can be used to reduce the emissivity. CoO has the highest absorptivity and higher absorption–emission ratio, indicating that its absorption performance is relatively good. Therefore, Ni and CoO were composed to prepare the solar selective absorption coating.

3 Calculation of optical properties of Ni–CoO composite coatings

3.1 Ni–CoO composite model construction and optical performance calculation

This study simulates the Ni–CoO composites that can be used to prepare the solar selective absorption coatings. The optimal ratio of Ni and CoO in the composite was studied to make the coating have the best optical performance.

First, the mixtures of Ni and CoO with different proportions were constructed, as shown in Table 3. Then, the Amorphous Cell module in Materials Studio software was

Table 2: Optical properties of Ni, Mo, CoO, and Cr_2O_3

Material	Absorptivity (α)	Emissivity (ε)	Absorption–emission ratio (α/ε)
Ni	0.354772	0.288738	1.228699
Mo	0.424343	0.355461	1.193782
CoO	0.617593	0.505377	1.222044
Cr_2O_3	0.270165	0.421665	0.640710

Table 3: Samples of simulation

Sample	m (Ni): m (CoO)	Number of Ni cells	Number of CoO cells	Volume ratio of Ni	Volume ratio of CoO
1	3:7	1	2	0.237	0.763
2	4:6	4	5	0.326	0.674
3	5:5	1	1	0.420	0.580
4	6:4	2	1	0.521	0.479
5	7:3	3	1	0.628	0.372
6	8:2	5	1	0.743	0.257

used to construct Ni–CoO molecular simulation systems with different ratios. The structure is shown in Figure 5. Then, we used the CASTEP module of Materials Studio software to calculate the absorptivities and emissivities of the composite materials. In this simulation, we selected the medium accuracy, so that the calculation has a shorter time and higher accuracy. And then, the reflectivities data of the composite models were derived. According to formulas (1)–(3), the absorptivity, emissivity, and absorption–

emission ratio of each sample model could be calculated, as shown in Table 4.

It can be seen from Table 4 that the absorption–emission ratio is largest when the mass ratio of Ni and CoO is 5:5. So, the single-layer Ni–CoO composite coating has the best optical performance when the mass ratio of Ni and CoO is around 1:1.

3.2 Validation of absorption and emissivity of composites

From Table 4, it can be found that the overall trend of the absorptivity and emissivity of the composite model becomes smaller as the Ni mass fraction increases, which is closer to the optical performance of Ni. It is also in line with the actual situation.

The equivalent medium theory [39] is a theory that specializes in metal-dielectric composite materials. When the two materials are mixed uniformly on the macro level

Figure 5: Composite structures of Ni–CoO in different proportions: (a) sample 1, (b) sample 2, (c) sample 3, (d) sample 4, (e) sample 5, and (f) sample 6.

Table 4: Optical properties of Ni–CoO composites

Sample	$m(\text{Ni}):m(\text{CoO})$	α	ϵ	α/ϵ
1	3:7	0.177835	0.202189	0.879548
2	4:6	0.165545	0.222805	0.743004
3	5:5	0.180313	0.200407	0.899734
4	6:4	0.168430	0.206079	0.817308
5	7:3	0.135460	0.193235	0.701012
6	8:2	0.094225	0.140272	0.671728

and non-uniform on the micro level, although the medium distribution and particle size are different in practice, the dielectric constant of the mixture can be equivalently calculated using some properties of these two materials.

The dielectric constant of composite materials was recorded as follows:

$$\tilde{\epsilon} = \tilde{\epsilon}_1 + i\tilde{\epsilon}_2. \quad (4)$$

The reflectivity is

$$R = \frac{[(\tilde{\epsilon}_1 + \sqrt{\tilde{\epsilon}_1^2 + \tilde{\epsilon}_2^2})^{1/2} - \sqrt{2}]^2 + (\sqrt{\tilde{\epsilon}_1^2 + \tilde{\epsilon}_2^2} - \tilde{\epsilon}_1)}{[(\tilde{\epsilon}_1 + \sqrt{\tilde{\epsilon}_1^2 + \tilde{\epsilon}_2^2})^{1/2} + \sqrt{2}]^2 + (\sqrt{\tilde{\epsilon}_1^2 + \tilde{\epsilon}_2^2} - \tilde{\epsilon}_1)} \quad (5)$$

Fang [40] used the equivalent medium theory to calculate the dielectric constants of Ni–CoO with different volume ratios and then calculated the reflectivities according to formulas (4) and (5). She further calculated the absorption–emission ratios of Ni–CoO with different volume ratios. She got the result that the absorption performance of the Ni–CoO composite is the best when the volume ratio of Ni and CoO is around 4:6.

It is guessed that the difference between the simulated data and the actual resulting from the force field, simulation time, size, and density of the system. What's more, the absorption–emission ratio was originally a small value, so the absorption–emission ratios of the composites in our simulation are smaller than that of the single substance. But the six composite models are calculated under the same force field, simulation time, system size, and density, the absorption–emission ratios of these six composite models are comparable. So, it can be concluded that which mass ratio of the Ni–CoO composite material has the best optical properties. This simulation calculation shows that the absorption–emission ratio is the largest when $m(\text{Ni}):m(\text{CoO}) = 5:5$. According to Table 3, we can get the volume ratio of Ni and CoO is 4.2:5.8 at this time. Our simulation result is in good agreement with the published result, which verified the availability of the simulation method. And we hope that our

method will provide some help for the study of solar selective absorption composite coatings.

4 Conclusion

To obtain a solar selective absorption composite film with good optical performance, we want to use a simple simulation method to replace the experiment to explore the optical performance of the composite material. In this paper, the density functional theory of the first principle was used to calculate the composite material that can be used to improve the optical properties of selective absorption coatings through Materials Studio software. The conclusions are as follows:

The reflectance curves of four materials, Ni, Mo, CoO, and Cr_2O_3 , were calculated, which showed the reflectance of each material for photons of different energy. Based on the standard AM1.5 of sunlight irradiance, the absorptivities of four substances were calculated using the integral formula. The absorptivities of these substances have a relationship of $\text{CoO} > \text{Mo} > \text{Ni} > \text{Cr}_2\text{O}_3$. The emissivity of each material was calculated according to the Planck formula and the integral formula of the emissivity, and the relationship was $\text{CoO} > \text{Cr}_2\text{O}_3 > \text{Mo} > \text{Ni}$. The absorption–emission ratio of each substance was obtained, and the relationship was $\text{Ni} > \text{CoO} > \text{Mo} > \text{Cr}_2\text{O}_3$. This is consistent with the conclusion that the absorptivities of Ni and Mo will not be very large obtained by analyzing their band structures and density of states. Based on the above indicators, Ni and CoO were selected to prepare the composite coating.

Approximately, the thickness of the coating has little effect on its optical performance, so the calculation and analysis are only made for the proportion problem. By constructing the composite models and analyzing their optical properties, the absorptivities, emissivities, and absorption–emission ratios of Ni and CoO at different ratios were obtained. It was found that the absorption performance was best when the mass ratio was 1:1. By comparing with experimental conclusions, the usability of the model is verified. And we hope that our method will provide some help for the study of solar selective absorption composite coatings.

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