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Experimental and numerical study on structural and thermal radiation properties of yttrium oxide sputtered on sapphire

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ABSTRACT

Materials for infrared domes are required to maintain good properties in harsh environments including mechanical strength, optical transmittance over a wide range of wavelengths and low emissivity. The purpose of this work is to sputter Y_2O_3 film onto a sapphire substrate by a radio frequency magnetron sputtering method and investigate the structural and thermal radiation properties of the films. In addition, the apparent emissivity of the coated sapphire is simulated for different film thicknesses at different temperatures. The experimental results show that the surface of the Y_2O_3 film is homogeneous, has a dense morphology and is totally polycrystalline. After being coated with the Y_2O_3 film, the transmission of the sapphire substrate is improved and the emissivity is decreased with increasing film thickness, especially at high temperatures. Simulation results show that apparent emissivity of a sapphire substrate at high temperatures can be reduced effectively by the Y_2O_3 film when the ratio of the thickness of Y_2O_3 film and sapphire substrate is larger than 0.01.

1. Introduction

The infrared dome is one of the key components of a hypersonic missile and must overcome thermal shock failure, withstand abrasion from raindrops and sand particles and provide maximum and precise signal transmission [1,2]. Therefore, the selection of the material for the infrared dome is very important. Due to the excellent optical and mechanical properties, sapphire is widely used for high speed domes for the medium-wave infrared, 3–5 μm [3]. However, the mechanical strength and transmission of sapphire decreases rapidly with increasing temperature [4], and the emissivity of sapphire increases sharply at elevated temperatures, which will lead to self-thermal radiation interference of the dome. Hence, it is very necessary improve its properties [5,6].

In recent years, there have been many studies on the methods to increase the flexural strength of sapphire at high temperatures, such as ion implantation [7], thermal annealing [8], neutron irradiation [9] and coating with protective films [10]. Demaree et al. [11] investigated the mechanical benefit of using highly-polished single-crystal *c*-axis sapphire implanted with Cr^+, Ti^+ and Si^+ ions both at room temperature and at 800 °C. Feng et al. [12] presented the results that at 600 °C and 800 °C, SiO_2 coatings increase the flexural strengths of *c*-axis sapphire by factors of 1.2 and 1.5, respectively. However, there is less research on methods to decrease the emissivity of sapphire at a high temperature.

Y_2O_3 is an important material for optical applications because of its excellent optical and mechanical properties including a wide transmission window from 250 nm to about 8 μm [13,14]. Y_2O_3 films have been deposited and characterized using many techniques for different purposes [15,16]. Zhang et al. [17] found that the hardness of ZnS coated with Y_2O_3 and SiO_2 multilayer increased from 250 to 430 kg/mm^2 and the transmittance increased from 63% to 83% at 3–5 μm. Rivera et al. [18] researched Eu:Y_2O_3 films on Si(100)/Si(111) substrates showing that they can improve the red emission by annealing the films in an oxygen atmosphere. Meanwhile, the emissivity of Y_2O_3 has low emissivity for the entire 3–5 μm band and insensitivity to high temperature effects [19]. Therefore, by being coated with an Y_2O_3 film, the emissivity of sapphire will decrease at high temperatures.

In this work, Y_2O_3 films were prepared on sapphire by a radio frequency magnetron sputtering method to improve the transmission and reduce the emissivity of sapphire. Subsequently, the apparent emissivity of the coated sapphire was simulated for different film thicknesses at different temperatures. The experimental results of structural and thermal radiation properties at different temperatures of the coated and uncoated sapphire are presented. The simulated apparent emissivity results are discussed in detail.

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2. Experimental

2.1. Preparation of the films

Y_2O_3 films were deposited on a sapphire substrate by a radio frequency magnetron sputtering method. After cleaning, the sapphire substrate was put into the deposition chamber on the specimen holder under the yttrium target (99.99%). The diameter and thickness of the substrate were 48 mm and 3 mm, respectively. The target diameter and thickness were 45 mm and 0.4 mm, respectively. The distance between the substrate and the target was 70 mm. The sputtering pressure and temperature were about 0.5 Pa and 600 °C, respectively. The substrate was first cleaned by Ar^+ for 15 min. Then re-sputtering and pre-sputtering were both carried out for 15 min. The sputtering power was 130 W, while the flow rates of the working gas (Ar) and the reaction gas (O_2) were 100 sccm and 4 sccm, respectively. The film thickness δ was determined by the deposition time and the thicknesses of film in this work were 1 μm and 3 μm. The sputtering conditions are listed in Table 1.

2.2. Characterization of the film

The structures of the Y_2O_3 films were analyzed by X-ray diffraction (XRD) using a Rigaku D/max-RB diffractometer with monochromated Cu Ka radiation at 40 kV and 40 mA. The plan-view and cross-section of the substrate-attached films were observed using a scanning electron microscope (SEM, Helios Nanolab600i). The composition of the film was determined to be Y_2O_3 by energy dispersive X-ray spectrometry (EDX, Helios Nanolab600i). The temperature-dependent optical transmission and emissivity were obtained by a Fourier Transform Infrared Spectrometer based on the energy method.

3. Numerical simulation on apparent emissivity

3.1. Optical parameters

For optical materials, the complex index of refraction $\hat{n}(v,T)$ is given below: [20]

$$\hat{n}(v,T)=\sqrt{\varepsilon(v,T)}=n(v,T)-\kappa(v,T)i \tag{1}$$

where $\varepsilon(v,T)$ is the complex permittivity; v is the wavenumber (reciprocal wavelength); T is the temperature; n is the index of refraction, and κ is the index of extinction.

The complex permittivity is

$$\varepsilon(v)=n^2-\kappa^2-2n\kappa i=\varepsilon_1-\varepsilon_2 i \tag{2}$$

The absorption coefficient $\alpha(v,T)$ is

$$\alpha(v,T)=4\pi v\kappa(v,T) \tag{3}$$

The reflectance R is [21]

$$R=\frac{(n-1)^2+\kappa^2}{(n+1)^2+\kappa^2} \tag{4}$$

Based on the classical pole-fit model, the lattice vibrations can be expressed as follows [22]:

$$\varepsilon(v,T)=\varepsilon_\infty+\sum_j\frac{\Delta\varepsilon_j(T)v_j^2(T)}{v_j^2(T)-v^2+\gamma_j(v,T)vi} \tag{5}$$

where $\Delta\varepsilon_j$, γ_j and v_j are the mode strength, line width and long-wavelength transverse optical frequency, respectively. ε_∞ is the high frequency permittivity.

Table 1
Sputtering conditions of Y_2O_3 film.

Conditions	Units	
Target	–	Y (99.99%)
Substrate	–	Sapphire
Ar flow rate	sccm	100
O_2 flow rate	sccm	4
Sputtering pressure	Pa	0.5
Substrate temperature	°C	600
Power	W	130
Substrate-to-target distance	m	0.07

The frequency dependence of γ_j is represented by the formula [23]

$$\gamma_j(v,T)=\gamma_j(T)\left\{\begin{matrix}1; & v\leqslant\bar{v}\\ \exp\left\{-\alpha\left[\left(\frac{v}{\bar{v}}\right)^2-1\right]\right\}; & v>\bar{v}\end{matrix}\right\} \tag{6}$$

where α and $\bar{v}$ are arbitrary parameters. It is proved that the model is successful when $\alpha = 4$ and $\bar{v}$ is 1.1 times the highest allowed infrared transverse optical mode frequency.

The temperature dependence of $\Delta\varepsilon_j$ and γ_j are described by the equations [24]:

$$\Delta\varepsilon_j(T)=\Delta\varepsilon_j(T_0)+b_j(T-T_0) \tag{7}$$

$$\frac{\gamma_j}{\gamma_{TOj}}(T)=\frac{\gamma_j}{\gamma_{TOj}}(T_0)+c_j(T-T_0)+d_j(T-T_0)^2 \tag{8}$$

$$\gamma_{TOj}(T)=\gamma_{TOj}(T_0)-a_j(T-T_0) \tag{9}$$

where T_0 is a reference temperature. a_j, b_j, c_j and d_j are constant coefficients depended by mode and material.

3.2. Apparent emissivity

The radiation characteristic of the semitransparent film is determined by the inner radiation of the film and the radiation of the interface between the film and the substrate. The apparent emissivity of the film is defined as the ratio of radiation of the film surface to the blackbody radiation of the surface at the same temperature. As shown in Fig. 1, a one-dimensional model is used to simulate the apparent emissivity because the thickness is far less than the size of the other dimensions. The emissivity of the substrate is ε_1. The absorption coefficient and refractive index of the film are k_a and n_2, respectively. While n_3 is the refractive index of air and the value is 1. The film is divided into N layers and the temperature of film T is uniform.

According to the Snell's law and energy conservation, when the ray travels from the film to the air, the radiation intensity at the interface is as shown below: [25]

$$(1-\rho_{23})\frac{I_2}{n_2^2}=\frac{I_3}{n_3^2} \tag{10}$$

where I_2 and I_3 are the radiation intensities on the left and right sides of the film surface; ρ_{23} is the Fresnel reflectivity when the ray travels from the film to the air and is expressed as:

$$\rho_{23}=0.5(R_S+R_P) \tag{11}$$

where, $$R_S=\left\{\frac{\left(\frac{n_3}{n_2}\right)^2\cos\theta-\left[\left(\frac{n_3}{n_1}\right)^2-\sin^2\theta\right]^{\frac{1}{2}}}{\left(\frac{n_3}{n_2}\right)^2\cos\theta+\left[\left(\frac{n_3}{n_1}\right)^2-\sin^2\theta\right]^{\frac{1}{2}}}\right\}^2,$$

$$R_P=\left\{\frac{\left[\left(\frac{n_3}{n_2}\right)^2-\sin^2\theta\right]^{\frac{1}{2}}-\cos\theta}{\left[\left(\frac{n_3}{n_2}\right)^2-\sin^2\theta\right]^{\frac{1}{2}}+\cos\theta}\right\}^2.$$

where θ is the incident angle. When $\theta = 0$, the ρ_{23} is

$$\rho_{23}=\left(\frac{n_3-n_2}{n_3+n_2}\right)^2 \tag{12}$$

Due to the film temperature being uniform, the radiant power emitted by each layer element is written as:

$$J=4n_2^2k_a\Delta xE_{b,k} \tag{13}$$