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Long-wave infrared emissivity characterization of vanadium dioxide-based multilayer structure on silicon substrate by temperature-dependent radiometric measurements

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ABSTRACT

This paper studies the IR properties of a VO_2-based multilayer structure with an emittance that increase with the temperature. A good tunability of the emissivity in long-wave infrared spectral region (8–12 µm) has been detected, with a positive emissivity differential of about 0.2. The transition of the long wave emissivity ε with the temperature is fully reversible according to a hysteresis cycle, with a transition temperature of 67 °C and a thermal bandwidth of only 8 °C. The multilayer structure also shows two emission peaks both in the heating cycle and in the cooling cycle. This emittance performance is promising for the future development of passive thermal control systems of spacecrafts.

1. Introduction

The thermochromic approach to control the internal temperature of the satellite is based on using specific coatings, called smart radiator devices (SRDs), with an emittance that can be tailored with the temperature [1–3]. Employing phase change materials with thermochromic properties in a SRD, such as vanadium dioxide, achieves this goal. Indeed, the radiant power emitted by surface unit in half space (W), depends on both surface temperature and object emissivity, according to the Stefan-Boltzmann law:

$$W = \varepsilon\sigma T^4, \quad (1)$$

where σ is the Stefan-Boltzmann constant, ε is the total emissivity, and T is the absolute temperature. Because the heat exchange between the spacecraft and the space environment is carried out mainly through radiation, an SRD can be mounted simply on the surface of the spacecraft and tailor the inside temperature of the spacecraft by adapting its emissivity (ε) to radiate more or less heat according to the change in orbit conditions.

Phase change materials, in particular transition metal oxides, present very promising electrical and optical properties for many applications in electronic and optical devices. Among transition metal oxides, vanadium dioxide (VO_2) is a particularly interesting material for SRD's thin film, because it has significant changes in electrical and optical properties due to external excitation such as temperature, electric field and/or optical signals [4–8]. Vanadium dioxide is characterized by a first order phase transition, and has a low temperature monocline structure in the semiconductor phase and a tetragonal rutile structure at high temperature in the metal phase [9,10]. A drastic increase in electrical conductivity, even three or four orders of magnitude, and strong variations in optical properties in the infrared region are observed above the transition temperature (T_C) of about 67 °C [11,12]. Numerous investigations have also been conducted to reduce the transition temperature, doping with metals such as tungsten, molybdenum and niobium have produced positive results in this direction [13]. However, despite the high potential of VO_2 for the SRD application, it is still difficult to reach a wide dynamic range (i.e., Δε tunability) for the appropriate emittance switching behavior with temperature in the mid-infrared range, along with the requirement to maintain sufficiently narrow the amplitude of the thermal hysteresis band [14]. In fact, since an SRD must have a low emissivity (ε_L) at low temperatures to maintain heat and a high emissivity (ε_H) at high temperatures to dissipate the additional heat that is not necessary, therefore, in terms of emissivity, the behavior suitable for SRD applications corresponds to an increase in the emission with the rise in temperature, contrary to what is generally desired to obtain in other application areas, such as in the infrared camouflage [15]. In order to maintain heat at low temperatures it is also important that the width of the hysteresis cycle is kept sufficiently tight.

In the present paper, we analyze the functional emissivity behavior for SRD applications as positive emissivity switching since

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$\Delta\varepsilon = \varepsilon_H - \varepsilon_L > 0$ [16]. Therefore, based on the previous considerations we use three evaluation parameters:

- *transition temperature* T_C, given by the temperature corresponding to the maximum of the emissivity derivative with respect to the temperature $\frac{\partial\varepsilon}{\partial T}\Big|_{max}$ and thus indicating the maximum induced emissivity variation by semiconductor-metal transition (heating) or metal-semiconductor (cooling);
- *thermal bandwidth* ΔT_C, given by the difference $T_{CH} - T_{CC}$ between the T_C of the heating cycle (T_{CH}) and that of the cooling cycle (T_{CC});
- *differential emissivity* $\Delta\varepsilon_{HL}$, given by the difference $\varepsilon_H - \varepsilon_L$ between the high-temperature (ε_H @100 °C) and low-temperature (ε_L @30 °C) emissivity values respectively corresponding to the metal and semiconductor phase of vanadium dioxide.

The radiometric characterization of the vanadium dioxide-based multilayer structure has been then performed through a temperature scanning of the sample from 30 °C to 100 °C and revealing the optical response of the structure to the long-wave infrared region (LWIR, 8–12 µm), where the main contribution of the thermal emission according to the laws of Planck and Wien is expected [17].

2. Material and methods

2.1. Sample description

The tested sample (Fig. 1) is a VO_2-based multilayer structure on a 480 µm thick polished-mirror crystalline silicon substrate.

The inner layer of VO_2 above the silicon is 230 nm, a thin copper film of 10 nm is deposited thereon and then another VO_2-layer of 450 nm is deposited. All layers were deposited by radiofrequency sputtering at a substrate temperature of 500 °C. As shown in the SEM image (Fig. 2a), the inner layer of VO_2 (in contact with Si) shows a more granular structure, while the upper layer is characterized by a more lamellar structure. The Cu layer being very thin is not evident in the SEM picture, but its presence has been confirmed by microanalysis (Fig. 2b).

The IR transparent silicon substrate has been chosen with the aim of having an almost zero emissivity structure for low temperatures. The sample has been designed and realised on the basis of a previous theoretical work studying the optimization of the emission properties in the mid-infrared of multilayer structures based on VO_2 [18].

In that work the tunability of the emissivity for $VO_2/Cu/VO_2$ multilayer structures was larger than the one found for $VO_2/Ag/VO_2$, as results from the numerical simulations based on the transfer matrix method, thus justifying the choose of copper as intermediate layer.

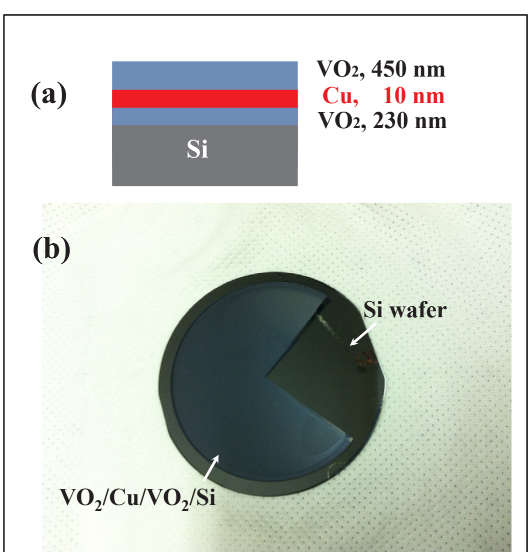
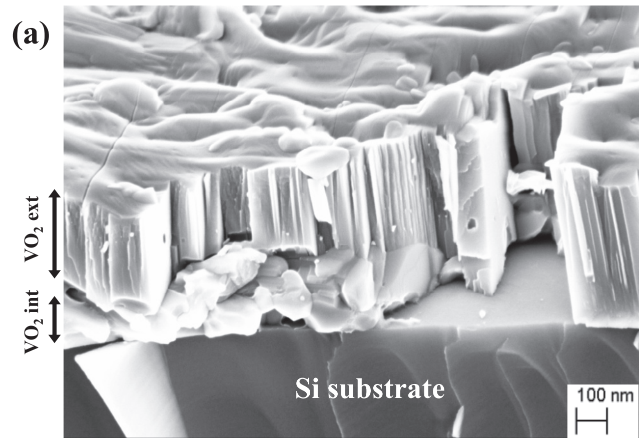


Fig. 1. (a) Sample structure; (b) sample image.



(b)

El	AN	Series	Net	unn. C [wt.%]	norm. C [wt.%]	Atom. C [at.%]	Error (1 Sigma) [wt.%]
V	23	K-series	1638	3.35	56.09	30.92	0.16
O	8	K-series	410	2.26	37.84	66.40	0.70
Cu	29	K-series	49	0.36	6.07	2.68	0.08
Si	14	K-series	143514	0.00	0.00	0.00	0.00
			Total:	5.98	100.00	100.00	

Fig. 2. (a) SEM image of the sample examined and (b) microanalysis.

2.2. Experimental setup

In order to evaluate the variation induced by the phase transition of VO_2 on the radiative emission, IR reflectance and transmittance measurements were performed as a function of the temperature of sample. In this way we can estimate the emissivity through Kirchhoff's thermal radiation law, which consists of equality between spectral emissivity and spectral absorptivity in thermodynamic equilibrium for a body:

$$\varepsilon_\lambda = \alpha_\lambda = 1 - r_\lambda - \tau_\lambda \tag{2}$$

where ε_λ is the spectral emissivity, α_λ the spectral absorptivity, r_λ and τ_λ the spectral reflectivity and transmissivity respectively.

In order to perform the temperature scanning measurements in the 30–100 °C range the sample was posed in contact with an electrical heater. A globar lamp was used as a source of infrared radiation and kept at a temperature of about 130 °C through a stabilized power supply. The radiation was modulated by a mechanical chopper (ORTEC 9479) before reaching the sample under test, and the same frequency is used for the external reference of the lock-in amplifier (Perkin-Elmer 7265) that provides data acquisition. For the reflectance measurements, performed at near-normal incidence, the IR radiation reflected by the sample was detected by a (HgCdZn)Te photovoltaic IR detector (Vigo System model PVI-4TE-10.6, quadrant cells 1 × 1 mm^2) with a rather flat responsivity in the MIR range (3–12 µm) coupled with a long pass filter with transmission of 90% in the LWIR (8–12 µm) range (see Fig. 3). In order to perform the transmittance measurements as a function of sample temperature, a central hole of 10 mm in diameter has been realized in the copper body of the electrical heater used for the sample temperature scan. The measurements were carried out at normal incidence of infrared radiation. The current temperature of the sample is measured by a copper-constantan thermocouple, and, in view of the low sample heat capacity, was used a type with thin and coated wires of 0.5 mm diameter (type TG-40-T, NY Thermoelectric Co., Inc.), in addition to a thermal flat with high thermal conductivity (RS Pro 554-311), consisting of metal oxide powders, to minimize the thermal resistance at the point of contact with the sample; and therefore the