Eur. Phys. J. Appl. Phys. **88**, 30301 (2019)

https://doi.org/10.1051/epjap/2020190339

THE EUROPEAN PHYSICAL JOURNAL APPLIED PHYSICS

Regular Article

Enhanced optical performance of thermochromic VO_2 based on multilayer designs

Houao Liu[1], Hao Song[1], Hao Xie[2], and Guanchao Yin[1,*]

[1] School of Materials Science and Technology, Wuhan University of Technology, Luoshi Road 122, Wuhan 430070, P.R. China
[2] School of Architecture and Materials Engineering, Hubei University of Education, High-tech Second Road 129, Wuhan 430205, P.R. China

Received: 27 November 2019 / Received in final form: 31 January 2020 / Accepted: 5 February 2020

Abstract. VO_2 is a widely studied thermochromic material for smart windows. In this work, we comprehensively studied optical responses of VO_2-based multilayer structures. It is discovered that one-layer antireflection layer is capable of effectively enhancing both luminous transmittance (T_{lum}) and solar transmittance (T_{sol}), solar spectrum modulation (ΔT_{sol}) is however quite moderate. Employing a two-top-layer strategy further improves the optical performance of VO_2, especially with an increase of ΔT_{sol} from 0.068 to 0.082. Remarkably, combining a layer with an index of 2.2 at the VO_2/glass interface continues to enhance the optical performance, leading to the highest T_{lum} and ΔT_{sol} among the investigated multilayer structures. Compared to the base structure of VO_2/glass, it contributes to a relative enhancement of 26.4% (from 0.435 to 0.550) for T_{lum} ($<\tau_c$), 35.3% (from 0.362 to 0.490) for T_{lum} ($>\tau_c$), and 71.7% (from 0.060 to 0.103) for ΔT_{sol}.

1 Introduction

Vanadium dioxide (VO_2) is a promising thermochromic material with a reversible phase transition upon temperature: it is monoclinic and permits infrared transmission below the critical temperature (τ_c, around 68 °C) while is metallic and blocks infrared transmission above τ_c [1–3]. This unique property offers VO_2 potentials to be widely applied in smart windows for energy saving and thus attracts great research interests in the last decade [1–8]. However, from optical aspect of view, VO_2 suffers from two issues limiting its wide application in large-scale architectural buildings. One is the relatively low luminous transmission (T_{lum}) originating from a severe absorption (Abs) and reflection (R) in the visible wavelength range, no matter which phase it is [9,10]. Another challenge is the poor solar spectrum modulation (ΔT_{sol}) during the phase transition process [9,10]. Simply increasing the thickness of VO_2 is capable of improving ΔT_{sol} to a certain degree but at a cost of deteriorating T_{lum} [5,11,12].

To enhance T_{lum} and ΔT_{sol} simultaneously, numerous efforts have been implemented mainly including multilayer designing [12–14], nanostructuring [11,15] and incorporating with other thermochromic materials [16,17]. Among the various strategies, multilayer designing is to incorporate one or more layers into the base VO_2/glass configuration, which modulates solar spectrum via Fabry-Perot interferences [12–14]. It is simple and quite feasible in the experiments. More importantly, the required fabrication methods are compatible with large-scale production in industry. A few specific multilayer stacks have been investigated in previous studies and success was achieved to various degrees [12–14]. However, previous investigations were mostly focusing on a fixed layer structure, lacking of systematic study of different multilayer configurations. Considering all factors analyzed above, in this contribution, we had limited the strategy to multilayer designing and systematically explored the potentials of various multilayer structures for achieving an optimum T_{lum} and ΔT_{sol} simultaneously.

2 Optical simulation

We intend to employ the software RefDex [18,19] for all optical parameters ($R/T/Abs$). It is programmed based on transfer-matrix method, which can deal with both coherent and incoherent propagation of light through multilayer systems with flat interfaces. Glass substrate was treated as an incoherent layer and the calculation of light propagation was done by the method as Harbecke suggested [20]. Optical constants of VO_2 are from reference [14], which are quite representative. Thickness of VO_2 is set as 50 nm, which is a reasonable value for tradeoff between T_{lum} and ΔT_{sol} [12–14].

To evaluate the visual and energy performance of VO_2-based multilayer structures, the integrated luminous transmittance (T_{lum}) and solar transmittance (T_{sol}) are

* e-mail: guanchao.yin@whut.edu.cn

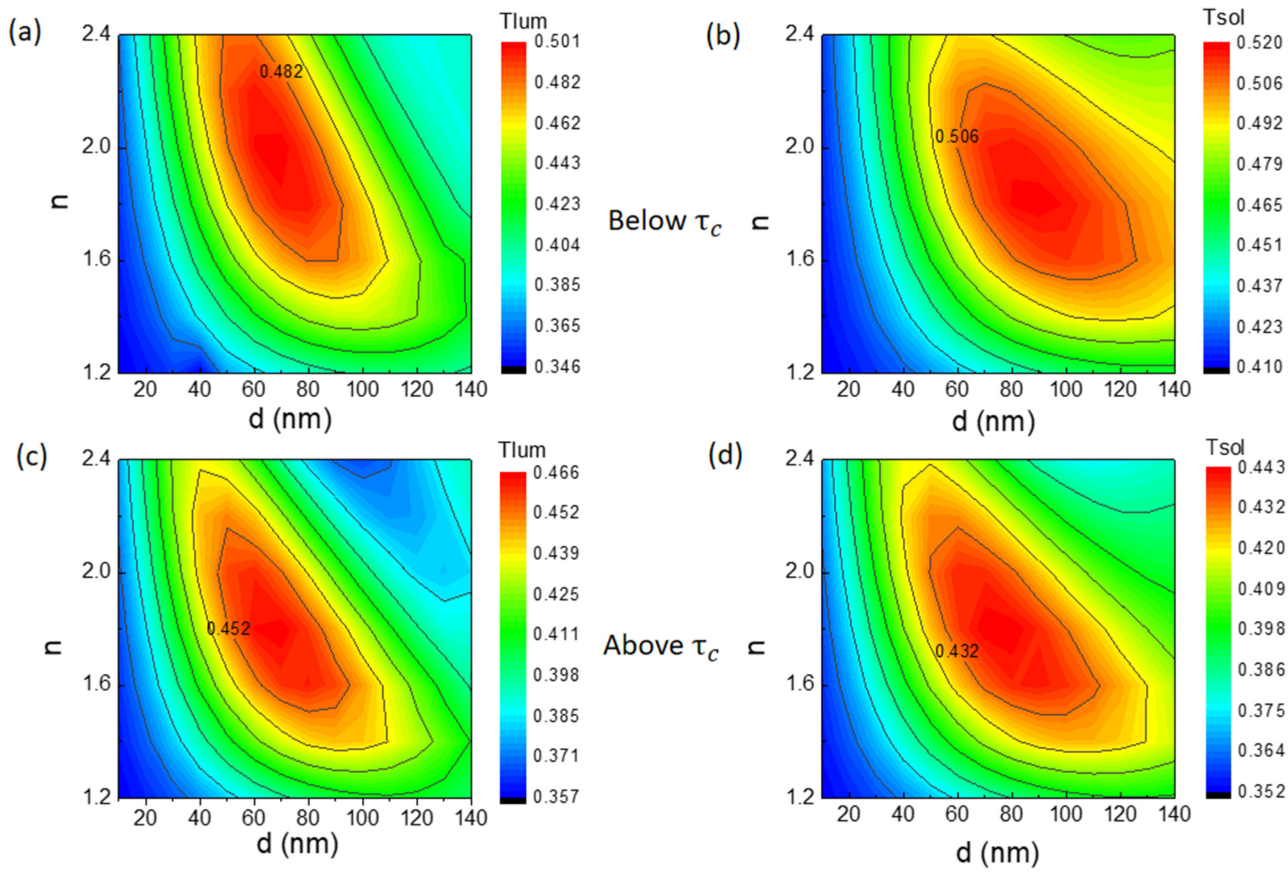

Fig. 1. T_{lum} and T_{sol} as a function of thickness (d) and refractive index (n) of the single top layer at both low (a, b) and high temperature (c, d).

defined as follows [16,17]:

$$T_{\mathrm{sol(lum)}} = \frac{\int \varphi_{\mathrm{sol(lum)}}(\lambda) T(\lambda) d\lambda}{\int T(\lambda) d\lambda} \tag{1}$$

where $\varphi(\lambda)$ is the solar spectrum under AM 1.5 solar irradiation condition, $T(\lambda)$ the calculated transmittance. Subscript *sol* indicates the wavelength range of 350–2500 nm and subscript *lum* the wavelength range of 380–780 nm.

To access the modulation degree of solar spectrum upon phase transition, solar spectrum modulation is simply denoted as shown in equation (2):

$$\Delta T_{\mathrm{sol}} = T_{\mathrm{sol}}(< \tau_c) - T_{\mathrm{sol}}(> \tau_c). \tag{2}$$

3 Results and discussion

3.1 One layer on top

We start from a single layer on top of the base VO_2/glass structure. To identify the reasonable layer on top, T_{lum} and T_{sol} values as a function of thickness (d) and refractive index (n) are plotted at both temperature conditions in Figure 1. It can be clearly observed that there exists an individual (n, d) zone, which corresponds to an optimized T_{lum} and T_{sol} in both temperature conditions. More elaborately, the four zones exhibit an overlap of (n, d), where the refractive index (n) and thickness (d) of the top layer were located in the range of around 1.6–2.2 and 60–90 nm, respectively. This is arising from that optical constants of VO_2 in the visible range between low and high temperature are similar [14]. The overlap of (n, d) indicates T_{lum} being capable of achieving optimized values at both temperature conditions with the same parameters for the top layer, which is desirable. Further, the broad (n, d) options offer great flexibilities to select materials for the top layer, which provides possibility to form a multifunctional windows with VO_2. For example, TiO_2 has an index of 2.2 and can offer an extra self-cleaning function [21].

T_{lum} and T_{sol} values for the top layer of ($n = 2.0$, $d = 80$ nm) are listed in Table 1 as representative values for the structure of layer 1/VO_2/glass. Compared to the pure VO_2/glass substrate, adding a top layer pronouncedly improves both T_{lum} and T_{sol}, from 0.435 to 0.498 for T_{lum} ($<\boldsymbol{\tau}_c$), 0.410 to 0.507 for T_{sol} ($<\boldsymbol{\tau}_c$), 0.362 to 0.461 for T_{lum} ($>\boldsymbol{\tau}_c$), 0.350 to 0.439 for T_{sol} ($>\boldsymbol{\tau}_c$). However, a top layer only yields to a ΔT_{sol} of 0.068, which is merely 0.008 higher than the base structure.

Table 1. Comparison of T_{lum} and T_{sol} for various multilayer structures.

	T_{lum} ($<\tau_c$)	T_{sol} ($<\tau_c$)	T_{lum} ($>\tau_c$)	T_{sol} ($>\tau_c$)	ΔT_{sol}
VO_2/glass	0.435	0.410	0.362	0.350	0.060
Layer 1/VO_2/glass	0.498	0.507	0.461	0.439	0.068
Layer 1/layer 2/VO_2/glass	0.501	0.549	0.455	0.467	0.082
Layer 1/layer 2/VO_2/layer 3/glass	0.550	0.609	0.490	0.506	0.103

Table 2. T_{lum} and T_{sol} at a (n, d) combination of top two layers (layer 1/layer 2/VO_2/glass).

Layer 1 (n_1, d_1)	Layer 2 (n_2, d_2)	T_{lum} ($<\tau_c$)	T_{sol} ($<\tau_c$)	T_{lum} ($>\tau_c$)	T_{sol} ($>\tau_c$)	ΔT_{sol}
(1.3, 113)	(1.7, 87)	0.459	0.524	0.441	0.445	0.079
(1.4,105)	(2.0, 75)	0.484	0.541	0.452	0.460	0.081
(1.5, 98)	(2.2, 66)	0.501	0.549	0.455	0.467	0.082
(1.6, 92)	(2.6, 58)	0.513	0.548	0.451	0.469	0.079
(1.7, 87)	(2.9, 51)	0.518	0.542	0.442	0.465	0.077
(1.8, 82)	(3.2, 46)	0.519	0.530	0.428	0.457	0.073

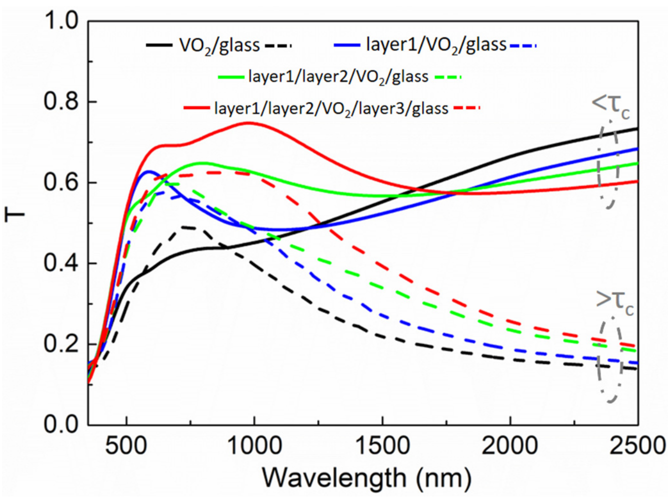

Fig. 2. T of various VO_2-based multilayer structures (black: VO_2/glass, blue: layer 1/VO_2/glass, green: layer 1/layer 2/VO_2/glass, red: layer 1/layer 2/VO_2/layer 3/glass).

Figure 2 plots R/T for both VO_2/glass (black) and layer 1/VO_2/glass (blue) structures. Above τ_c, T in the infrared range is pronouncedly reduced due to VO_2 transferring to metallic phase with high k (extinction coefficient) values in the long wavelengths for both structures. This is the typical optical feature of VO_2, which is agreeable with previous reports [16,17]. After the coating, T in the wavelength range of 380–780 nm is largely improved and thus contributes to a higher T_{lum}, which is mainly resulted from the reduction of R. Notably, typically a low-index material (such as SiO_2 and MgF_2 with 1.5 and 1.28, respectively) is preferred as an antireflection layer since it can effectively reduce R. Here, we realized that high index materials are more suitable for VO_2. Unfortunately, when the temperature is beyond τ_c the top antireflection layer increases T in infrared range at the same time, which is actually not desired for smart windows. This explains why ΔT_{sol} stays almost stable after coating the top layer.

3.2 Multilayer structures

It is widely studied that multilayer structures can induce better anti-reflection effects than a single layer since they can cover broader wavelength range [22,23]. To verify it, we investigate the structure of two top layers (layer 1/layer 2/VO_2/glass) and calculate T_{lum} and T_{sol}. According to the rule of W-type antireflection layers [24,25], Table 2 lists a few (n, d) combinations for optimized T_{lum} and T_{sol}. We can observe that T_{lum} ($<\tau_c$) exhibits an obvious enhancement only at the condition of Layer 1 with an index ≥1.5. Besides, at an index of 1.5 for Layer 1, this combination offers a maximum ΔT_{sol}. Refractive index of Layer 2 is required larger than that of Layer 1 and a higher refractive index of Layer 1 indicates an even higher one for Layer 2. This implies that if $n_1 > 1.5$, n_2 should be larger than 2.2, which is challenging to select a proper material for Layer 2 among the common materials. Considering the factors above, the (n, d) combination of (1.5, 98) and (2.2, 66) is preferred since the common materials of SiO_2 and TiO_2 have an index of 1.5 and 2.2, respectively. This is consistent with the experimentally reported SiO_2/TiO_2/VO_2 structures [12–14]. Therefore, in the following, we had confined ourselves to the combination of (1.5, 98) for Layer 1 and (2.2, 66) for Layer 2 for further discussion and the corresponding T_{lum} and T_{sol} are taken as representative values and presented in Table 1. Compared to the one-layer coating, the double-layer antireflection structure doesn't offer a significant improvement in T_{lum} or T_{sol}. Fortunately, ΔT_{sol} is as high as 0.082, which is 0.014 higher than that for the one-top-layer structure.

Looking closely VO_2 and glass substrate, it is realized that an obvious gap in refractive index still exist, which implies that the VO_2/glass interface reflection is not marginal. To suppress it, we inserted a layer into the VO_2/glass interface and form a structure of layer 1/layer 2/VO_2/layer 3/glass. Fixing the parameters of the top two layers as listed in Table 1, a layer of (2.2, 67) is found to be proper for Layer 3. Due to the inserting layer, the structure exhibits the most significant enhancement of T in the broad wavelength range of 600–1700 nm (red lines, Fig. 2). As a result, T_{lum} reaches as high as 0.550 at temperature $<\boldsymbol{\tau}_c$ and 0.490 at temperature $>\boldsymbol{\tau}_c$, which is a great improvement of 26.4% and 35.3% (compared to the base structure), respectively. Simultaneously, the layer 1/layer 2/VO_2/layer 3/glass structure also leads to a large ΔT_{sol} of 0.103.

4 Conclusions

In this work, we systematically investigated the impact of various VO_2-based multilayer structures on T_{lum} and T_{sol}. For the structure of layer 1/VO_2/glass, despite of bringing an obvious enhancement in T_{lum}, it fails to reduce the reflection in the infrared range and thus shows a moderate ΔT_{sol} of 0.060. Double anti-reflection layers can further improve optical performance of VO_2 with a ΔT_{sol} of 0.082. Remarkably, further inserting a layer with an index of 2.2 at the interface of VO_2/glass, the layer 1/layer 2/VO_2/layer 3/glass structure offers the highest T_{lum} values. Compared to the base structure of VO_2/glass, the relative enhancement is 26.4% for T_{lum} ($<\boldsymbol{\tau}_c$) and 35.3% for T_{lum} ($>\boldsymbol{\tau}_c$). Simultaneously, this structure yields to a ΔT_{sol} of 0.103, which is the optimum value among those investigated multilayer structures. Overall, for achieving a super optical performance of VO_2-based multilayer structures, the combination of antireflection layers on top and at the interface of VO_2/glass is therefore essential.

The authors would acknowledge the funding support of the Fundamental Research Funds for the Central Universities (WUT:183101002, 193201003), Hao Xie specially the support by science and technology planning project of Department of Housing and Urban-Rural Development of Hubei Province, China.

Author contribution statement

Houao Liu is the main contributor to the calculation and manuscript writing. Hao Song is responsible for the calculation below the critical temperature. Hao Xie offers the suggestions for parameter determination and Guanchao Yin takes the full responsibility for the whole work, including idea design, calculation guidance, manuscript corrections.

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Cite this article as: Houao Liu, Hao Song, Hao Xie, Guanchao Yin, Enhanced optical performance of thermochromic VO_2 based on multilayer designs, Eur. Phys. J. Appl. Phys. **88**, 30301 (2019)