VO₂ thin films for smart windows: Numerical study of the optical properties and performance improvement

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Abstract. Vanadium dioxide (VO₂) have attracted tremendous interest in recent years due to their semiconductor-metal transition near room temperature. In this work, we report numerical simulation of the optical properties of VO₂ thin layers which include the index of refraction, the coefficient of extinction, the reflectance and the transmittance. In this sense, we used the Model of Drude-Lorentz for calculating coefficients at from the optical parameters n and k using the determination of the dielectric constant ε (ω, T). Knowing that the semiconductor-metal transition temperature of VO₂ is 68 °C, the thickness effect on the optical properties of VO₂ thin films has been studied by transmittance simulations. Our results revealed a significant changes in optical properties of VO₂ thin layers, which produces many interesting applications especially for smart windows.

1. Introduction
Vanadium dioxide "VO₂" has a semiconductor-metal phase transition at about ~ 68 °C, resulting in significant changes in electrical resistivity and its optical IR properties as a result of its strong electrons correlation [1,2].

The behavior of the band structure of VO₂ changes as a function of temperature [3]. From theoretical point of view, this phase transition has been, initially, interpreted in terms of Mott-Hubbard transition [4]. More precisely, in the metallic state, above the transition temperature, the formation of VO₂ in phase (R) destroys the V-V metal bonds, resulting in the absence of the band gap. At the same time, the overlap of the conduction and valence band facilitates the displacement of electrons forming electron-hole pairs (figure 1.a). This appearance of charge carriers in VO₂ would optically explain the decrease in transmittance and the increase in reflectance in the metallic state of VO₂. In the semiconductor state, as a result of the formation of strong V-V metal bonds, the symmetry is broken and the structure of VO₂ is changed to monoclinic phase M. These metallic bonds are the cause of the separation of the d|| band (figure 1.b) and the birth of the band gap (E₉)
which has been determined to be 0.6 eV [5,6]. As a result of modifications in the electronic and electrical properties of VO$_2$ during the phase transition, the optical properties change, for which VO$_2$ is transparent in the hyper frequency domain in the semiconductor state (T < 68°C), and becomes reflective and opaque in the same wavelength range in the metallic state (T > 68°C). Due to its optical transition properties, VO$_2$ has been developed for innovative applications in various fields, particularly in smart windows [7-9].

In this work, we used the Drude-Lorentz model to calculate the optical properties of semiconductor and metallic VO$_2$ thin films that include the refractive index (n), extinction coefficient (k), reflectance (R) and transmittance (T). The thickness effect on the optical properties of VO$_2$ thin films has been studied by transmittance simulations, revealing dramatic changes in the optical properties of VO$_2$ thin films, which produces many interesting applications especially for smart windows.

2. General formalism

Using the classical dispersion model based on the sum of Lorentz and Drude oscillators, we can calculate the optical constants of VO$_2$ thin films in function of the frequency. Furthermore, the dielectric functions of the VO$_2$ films were calculated by taking into consideration the parameters of VO$_2$ film reported by Kana Kana et al [10].

$$\varepsilon(\omega, T) = (n(\omega, T) + ik(\omega, T))^2$$  \hspace{1cm} (1)

These two constants n and k represent respectively the real and imaginary part of the complex dielectric constant $\varepsilon$ of VO$_2$.

$$\varepsilon(\omega, T) = \varepsilon_{\infty} + \varepsilon_1(\omega, T) + \varepsilon_2(\omega, T) + \varepsilon_3(\omega, T)$$  \hspace{1cm} (2)

$$\varepsilon(\omega, T) = \varepsilon_{\infty} + \frac{(\varepsilon_s - \varepsilon_{\infty}).\omega^2}{\omega_1^2 - \omega^2 + i\Gamma_0.\omega} + \sum_{i=1}^{N} \frac{f_i.\omega_{0j}^2}{\omega_{0j}^2 - \omega^2 + i\gamma_j.\omega} + \frac{\omega_p^2}{-\omega^2 + i\Gamma_d.\omega}$$  \hspace{1cm} (3)

![Diagram showing phase transition of VO$_2$](image)

**Eg = Eg(\lambda Temperature)**
In the semiconductor state, with the first three terms corresponding to the dispersion of the Lorentz model, the optical constants of VO$_2$ are adjusted. This formula is based on the classical theory of light-matter interaction, and is used also to describe the frequency-dependent polarization caused by the bound charges.

In the metallic state, at the dielectric function, the last term referring to the Drude model is added because in the metallic phase, VO$_2$ acts as a Drude metal with high absorption [11].

We simulated the interaction of an electromagnetic wave with a thin layer of VO$_2$ with a thickness of 82 nm [10]. For this purpose, we have developed a Matlab program to calculate the optical properties of semiconductor (30°C) and metallic (85 °C) VO$_2$ thin films for wavelengths range 380 nm to 2500 nm. We also investigate the role of film thickness on the optical transmittance.

3. Results and Discussion

3.1 Simulation of optical indices

The wavelength dispersions of the refractive index, n, and extinction coefficient, k, of VO$_2$ thin films, with a 82-nm-thick, (semiconductor at T=30 °C and metal at T=85 °C) over a spectral range covering the UV-VIS and partially NIR, more accurately over 300 – 2500 nm, are shown in figure 2.

![Figure 2](image_url)

Figure 2. (a) Refractive index, n and (b) extinction coefficient, k of VO$_2$ thin films (e = 82 nm) at two different temperatures; 30° and 85°C.

Figure 2 clearly shows that the switching of the optical constants n and k of VO$_2$ as the outside temperature increases for the near-infrared region is strongly accentuated in comparison with the visible region. The refractive index decreases by 55% at about $\lambda=1000$ nm; it is reduced from 3.2 to 1.6 between the semiconductor state (30 °C) and the metallic state (85 °C) respectively. According to the works by Kakiuchida et al. and Kana Kana et al. [12, 10]. Hence, this significant modification of the optical constants of VO$_2$ supports its excellent thermochromic properties.
3.2 Optical transmittance and reflectance simulations

Figure 3 illustrates the optical transmittance and reflectance of VO$_2$ thin films with a 82-nm-thick for two different temperatures; 30° and 85 °C, which are selected to cover the optical response between the IR transmission state and the opaque IR state. As we can see, the optical transmittance and reflectance exhibits a high temperature dependence in the infrared spectral region in which there is no significant change in the visible region.

At λ=2500 nm, the optical transmittance is approximately 70% and 20% below and above the transition temperature of $T_C \sim 68 °C$, this optical modulation transmittance decreases with increasing temperature [12,13].

The reflectance evolves as the temperature changes, considering the relationship $R = 1- T$, the optical reflectance is reduced by 66% at λ=2500 nm; it is reduced from 70 % to 22 % between the metallic state (85 °C) and the semiconductor state (30 °C) respectively.

![Figure 3](image_url)

**Figure 3.** Optical (a) transmittance and (b) reflectance of VO$_2$ thin films (e= 82 nm) in the spectral range 300 - 2500 nm at temperatures below and above the phase transition temperature (68°C).

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3.3 Influence of the thickness

Figure 4 reports the optical transmission versus semiconductor-metallic phases of VO$_2$ thin films for three different thickness; 300, 600 and 900 nm. In the visible range, the optical transmission switching is smaller, whereas in the near infrared it is more than 40%; at 2500 nm, particularly, the optical modulation is approximately $\sim 52\%$.

The results of this numerical study show the evolution of the optical properties of vanadium dioxide as a function of the thickness. As can be observed, in the semiconductor state (32 °C), the optical transmittance increases from 70, 72 to 75 and 78%, at 1000 nm, with increased thickness from 82, 300 to 600 and 900 nm respectively, and there is no significant change in the partially FIR region. In the metallic state (85 °C), we observe an increase of the optical transmittance until 31%, with an increase of the thickness in the NIR and FIR regions, especially, the optical modulation is
about \( \sim 11\% \). Therefore, the optical transmittance increases slightly while increasing the thickness of \( \text{VO}_2 \) [14]. These results confirm the high potential of vanadium dioxide for smart window applications in the automotive industry and architectural construction (figure 4), this clear modulation of IR optical transmission to IR optical reflection can be applied for optical fibre-based devices [15].

![Graph showing optical transmittance profiles of VO₂ thin films.](image)

**Figure 4.** Optical transmittance profiles of \( \text{VO}_2 \) thin films (semiconductor at \( T=30^\circ\text{C} \) and metal at \( T=85^\circ\text{C} \)) for three different thickness; 300, 600 and 900 nm respectively.

4. Conclusion
This paper has presented a numerical study of the optical properties of \( \text{VO}_2 \) thin films, with a 82-nm-thick, deposited on a glass substrate. The optical constants of the film were calculated using the classical dispersion model based on the sum of Lorentz and Drude oscillators. These optical constants exhibit a high temperature dependence in the infrared spectral region in which there is no significant change in the visible region. As a consequence, this significant modification of the optical constants of \( \text{VO}_2 \) supports its excellent thermochromic properties. Furthermore, the optical transmittance of different thicknesses was presented, in which we found that the optical transmittance of \( \text{VO}_2 \) thin film with 84 nm increases slightly while increasing the thickness. Our results highlighted the improvement of the optical properties to the practical application of \( \text{VO}_2 \)-based smart windows.

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