Energy-saving potential of thermochromic coatings in transparent building envelope components

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Abstract

Advances in the energy management of buildings are essential for reducing the carbon footprint in the building sector. Applying special window coatings of varying optical properties offers new chances for improved energy efficiency. Thermochromic vanadium oxide (VO2) is an important material for this development and is, therefore, one of the most investigated thermochromic materials. It changes its transmittance in the infrared spectral range in response to a changing temperature. In this study, VO2 coating was deposited on ultra-thin flexible glass in a continuous roll-to-roll sputtering process. The thermochromic layer had a thickness of 70 nm, and it was embedded between two zirconium oxide layers of 170 nm each. The luminous transmittance of the stack was 50%. A solar modulation of 9.6% was reached between the low and high-temperature states. The transition temperature between the cold infrared transparent and the warm infrared opaque state was determined to be 22°C. Different application scenarios for this material were evaluated. The modulation of the solar transmittance was calculated for the combination of VO2 with state-of-the-art low-e coatings. Our findings show that such a combination does not offer a benefit for reducing the energy demand of a building. However, a stand-alone implementation of thermochromic coatings has a high potential if the energy consumption of the building is dominated by cooling demands.

Keywords

smart coatings, energy saving, radiative cooling, smart windows, electrochromic coatings, thermochromic coatings, energy efficiency, vanadium oxide

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1 INTRODUCTION

The building sector in Europe is presently responsible for roughly 40% of the primary energy consumption and 36% of the greenhouse gas emissions (D'Agostino & Mazzarella, 2019). In the Scandinavian countries and Central Europe, energy is mainly used for heating and illumination. In contrast, in Southern European countries, cooling and air conditioning are the main sources of energy consumption. Several global trends support the increased energy demand for cooling the interior of buildings: climate change leads to generally increased average temperatures. Additionally, the continuous growth of both the world population and the welfare in southern countries causes an effect in the same direction. Transparent components of the buildings, i.e. windows and skylights, are crucial for the energy exchange between the interior and the surroundings. This is especially significant considering the increased window-to-wall ratio (WWR) of non-residential buildings over the last decades.

Since the 1970s, considerable improvements have been made to intentionally control the energy flux through windows. During this time, low emissivity (low-e) coatings became popular. They reduce the heat loss to the outside by thermal radiation. In contrast, solar control coatings prevent the near-infrared radiation of the sun from entering the building. These types of coatings decrease the energy demand for cooling on days with intense sunshine (Yaşar & Kalfa, 2012), (Teixeira et al., 2020). Nowadays, all these approaches are considered to be state-of-the-art. The heat exchange through the window is specified by its U-value (degree to which a building component prevents heat from transmitting between the inside and the outside of a building) and the g-factor (total solar transmittance of a window, characterizing the heat gain at sunshine conditions). Details about the definition of these values can be found in (Jelle, 2013) and (ISO 9050, EN 673:2011).

Low-e and solar control coatings have static optical properties. The preferred version can be installed in a building depending on both the design specifics and the local climate. However, as mentioned above, the optical properties of these windows cannot be adapted as a response to daily or seasonal changes. Therefore, the need for further improvements in energy efficiency has drawn increasing attention to so-called smart materials. Coatings made with these materials can change their optical properties when triggered by an external stimulus.

There exist different types of smart materials. They can be distinguished by the type of stimulus which causes the change in the optical properties. The most common ones are electrochromic, gasochromic, photochromic, and thermochromic materials. Among these, the electrochromic approach is the most popular; it has already been realized in many installations. This type of smart coating has the advantage of a large switching effect in the visible spectral range. Moreover, it can be actively controlled by a user. The effect of windows with electrochromic coatings on the energy efficiency of buildings has already been extensively investigated (Baldassarri et al., 2016).

This article focuses on the energy-saving potential of thermochromic coatings. Thermochromic coatings change their properties in response to the temperature. Different materials with such properties are presented in the literature (Wang, 2021), (Crosby & Netravali, 2022). Vanadium dioxide (VO_2) is the most promising and well-investigated thermochromic material for energy-saving applications (Hu et al., 2023), (Wang et al., 2021). It undergoes a phase transition between a low-temperature monoclinic crystalline structure and a high-temperature tetragonal structure. This phase transition is accompanied by a change of the optical properties in the infrared spectral range, i.e. the low-temperature phase is transparent while the high-temperature phase is opaque. The optical properties in the UV and visible spectral ranges are nearly unaffected by this change.

The transition temperature of pure VO_2 is 68°C. Previous research has established that this value can be reduced to approximately 25-30°C by introducing small quantities of tungsten into the coating (Jin et al., 1998). However, the widespread usage of thermochromic vanadium oxide is still being prevented by various challenges (Chang et al., 2018). First, the switching effect is limited to the infrared spectral range. Thus, achievable energy modulation is lower compared to the electrochromic counterparts, which also change their properties in the visible spectral range. Secondly, the luminous transmittance of vanadium oxide thin films is only around 40%. Moreover, the absorption in the visible spectral range causes a yellowish colour of the transmitted radiation. Both aspects are undesirable optical properties which speak against the use of thermochromic coatings in windows. Furthermore, the deposition process of vanadium oxide is very demanding, as the phase diagram of the vanadium-oxygen system includes numerous other phases without thermochromic properties. Therefore, process parameters need precise adjustment to ensure the formation of the desired stoichiometry (Bahlawane & Lenoble, 2014). Different techniques have been investigated for the fabrication of thermochromic VO₂, among them chemical vapour deposition (Bahlawane et al., 2014), (Mohammad et al., 2023), hydrothermal methods (Magdassi et al., 2017), and physical vapour deposition (Vu et al., 2019), (Rezek et al., 2022).

This study addresses several of the challenges of VO₂. First, the VO₂ layer was embedded between two zirconium oxide (ZrO₂) layers to increase the optical performance as well as the environmental stability compared to the single-layer vanadium oxide. Second, an improved roll-to-roll deposition process was developed as a possible pathway to the low-cost manufacturing of thermochromic coatings. This system included an inline monitoring system for the transmittance of the coated material, which resulted in increased stability compared to a process version presented in a previous work (Rezek et al., 2022). Thin flexible glass was chosen as a substrate. This bendable material has an areal weight of less than 0.3 kg/m². Its light weight increases the suitability of the coated material for building retrofits, making the installation of thermochromic-coated materials relatively easy.

Preferably, smart coatings should be used in combination with well-established state-of-the-art solutions. Great effort has been put into answering the question of how the ability of adaptation can push the energy efficiency of buildings beyond the presently existing limits. Detailed analyses of the presently known effects on building level can be found in (Tällberg et al., 2016) and (Butt et al., 2021).

The methodology section of this article introduces the calculation scheme of different quantities. It is later used for the evaluation of the results. Based on the obtained results, different application scenarios for thermochromic coatings are evaluated. The coating process itself is only presented briefly since the deposition technique is not the focus of this paper.

2 METHODOLOGY

The thermochromic materials are characterized by measuring the optical properties across a temperature range of -20°C up to 80°C. The expected optical effect of vanadium oxide in a window is schematically shown in Figure 1.



FIG. 1 Effect of a thermochromic coating on the transmission of solar radiation through an integrated glass unit in the cold state (left) and warm state (right)

The transmittance of VO_2 in the visible spectral range remains nearly constant across the entire temperature range. The change in the optical properties is mainly observed in the near-infrared spectral range. Here, the transmission in the cold state is much higher than in the warm state.

The energy-saving potential can be evaluated based on both the measured luminous transmittance of the coated glass samples and the difference in the measured solar transmittance in the high- and low-temperature states. The definition of these quantities was taken from ISO 9050:2003.

The luminous transmittance T_{lum} is calculated as (1)

$$T_{lum} = \int_{380 nm}^{780 nm} T(\lambda) D(\lambda) V(\lambda) d\lambda / \int_{380 nm}^{780 nm} D(\lambda) V(\lambda) d\lambda$$
(1)

where $T(\lambda)$ is the transmittance spectrum of the coated glass, $D(\lambda)$ is the spectral distribution of illuminant D65, and $V(\lambda)$ is the spectral luminous efficiency for photopic vision.

The change in solar transmittance ΔT_{sol} is defined by (2) and (3) as the difference between the low-temperature and the high-temperature value measured at -20°C and 80°C, respectively.

$$\Delta T_{sol} = T_{sol,LT} - T_{sol,HT} \tag{2}$$

$$T_{sol,(LT/HT)} = \int_{300 nm}^{2500 nm} T(\lambda,\Theta) S(\lambda) d\lambda / \int_{300 nm}^{2500 nm} S(\lambda) d\lambda$$
(3)

Where $T(\lambda, \Theta)$ is the temperature-dependent transmission, and $S(\lambda)$ the relative spectral distribution of the solar radiation. Both T_{lum} and ΔT_{sol} are usually given in percentages.

For practical applications, it is important to know transition temperature Θ_{tr} between the high- and low-temperature phases. It would even be advantageous to adjust this value according to the climatic conditions of the intended installation. As already mentioned, tuning of Θ_{tr} is usually achieved by incorporating different amounts of tungsten into VO₂.(Jin et al. 1998). However, vanadium oxide shows a thermal hysteresis behaviour, i.e. the optical properties of the layers at a certain temperature are different depending on whether the system has been cooled down or heated up. In this work, the transition temperature Θ_{tr} is defined as the centre point of the hysteresis of the transmittance values measured at 2500 nm as proposed in (Houska et al., 2019).

Nevertheless, the materials reported in this paper exhibit a non-symmetrical hysteresis. Therefore, the definition of a centre point of the hysteresis must be refined to obtain reproducible values. The advanced calculation procedure of the transition temperature can be explained with the help of the quantities given in Figure 2.



FIG. 2 Definition of the transition temperature of a thermochromic system

The left side of Figure 2 shows a typical non-symmetric hysteresis of the transmission at 2500 nm. The right side of Figure 2 shows the difference between the upper value of the transmittance (obtained for the heating curve) and the lower value (obtained for the cooling curve) as a function of the temperature ($\Delta T(\Theta)$). There is a disctinct maximum in this curve which can be interpreted as the transition temperature Θ_{rr} .

An alternative approach defines the transition temperature as the abscissa value of the vertical line that divides the enclosed area in Figure 2 (left) into two equal parts. This can be expressed by the integral equation (4).

$$\int_{-20}^{\Theta_{tr}} \Delta T(\Theta) d\Theta = \frac{1}{2} \int_{-20}^{80} \Delta T(\Theta) d\Theta$$
⁽⁴⁾

For the example given in Figure 2 (left), $\Theta_{tr} = 25^{\circ}C$ is obtained for the maximum approach, and $\Theta_{tr} = 22^{\circ}C$ for the divided area approach. In the following, the divided area approach according to equation (4) will be applied if the authors refer to the transition temperature.

This choice takes all measurement points into consideration. Hence, it is less sensitive to deviations of single values. In the case of a symmetric hysteresis, both values are identical and equal to the value obtained by the acknowledged procedure described in (Houska et al., 2019).

A thermochromic three-layer system $ZrO_2-VO_2-ZrO_2$ was deposited on ultra-thin flexible glass. The approach followed the concept outlined in (Vlcek et al., 2017).

The substrate was ultrathin glass (NEG) with a width of 300 mm and a thickness of 0.1 mm. The flexibility of the substrate allowed the application of a roll-to-roll coating approach. Therefore, the experimental work was carried out in the roll-to-roll sputtering equipment FOSA labX (VON ARDENNE) (Figure 3).



FIG. 3 Schematic drawing (left) and picture (right) of the roll-to-roll sputtering equipment FOSA labX for the deposition of the Zr02-V02-Zr02 layer stack on ultra-thin glass

A roll of this material with a length of 20 meters was installed in the position of the pay-out roller. The material was spliced between 10-meter-long polymer films as leader and follower, respectively. This ensured complete protection of the coated material inside the roll during the evacuation and venting of the vacuum chamber. The material can be conveyed back and forth between the pay-out and take-up rollers. On its way, it passes two deposition zones. The left and right deposition positions in the scheme shown in Figure 3 were used for the deposition of the thermochromic vanadium oxide and the zirconium oxide top and bottom layers, respectively. In this type of machine, at least two passes of the substrate through the machine were necessary to deposit the complete three-layer stack.

The zirconium oxide layer was sputtered using a rotatable magnetron with a zirconium oxide ceramic target (GfE FREMAT). The length of the target was 650 mm. The material was sputtered at 6 kW pulsed DC power ($t_{on} = 16 \ \mu s$:, $t_{off} = 4 \ \mu s$). The dynamic deposition rate was 22 nm*m/min. The argon gas flow of 250 sccm was regulated by a mass flow controller (MKS instruments). The oxygen flow of 8 sccm was regulated by a second mass flow controller (MKS instruments). This was the minimum amount of oxygen to obtain a zirconium oxide layer with an extinction coefficient k<10⁻³. The chamber pressure was 0.4 Pa.

The vanadium oxide layer was sputtered using a rotatable magnetron with a vanadium-tungsten alloy target (atomic percentage of W: 1.2 at%, GfE FREMAT). This vanadium-to-tungsten ratio is well-known for shifting the transition temperature from 68° C to ambient values [14]. The length of the target was 650 mm. The material was sputtered at 6 kW using a high-power impulse magnetron sputtering (HiPIMS) power supply (nano4Energy). The duty cycle was 5.25% ($t_{on} = 70 \text{ ms}$: , $t_{off} = 1263 \text{ ms}$). The dynamic deposition rate was 11 nm*m/min. The glass was pre-heated to 350° C by a radiation heater (Figure 3). The argon gas flow of 250 sccm was regulated by a mass flow controller (MKS instruments). The oxygen flow was regulated by a second mass flow controller (MKS instruments). The layer properties are very sensitive to the amount of oxygen introduced. Details about the interdependence of coating parameters and thermochromic properties were previously reported (Rezek et al., 2022). Metallic samples were sputtered for oxygen flow values below 22 sccm. Oxygen flow values higher than 26 sccm provided vanadium oxide targets without thermochromic properties. An inline monitoring system for the transmittance was used to determine the correct range of oxygen flow (Figure 3). The control scheme of the process is shown in Figure 4.



FIG. 4 Schematic drawing of the control setup for the reactive sputtering of vanadium oxide

In the first pass, a zirconium oxide layer was sputtered onto the substrate. The thickness of this base layer was kept constant at 170 nm for all experiments. The vanadium oxide layer of 80 nm thickness was deposited in the second pass. The oxygen flow was adjusted to 25.4 sccm. This value was chosen based on the transmission inline monitoring value of 33% (at 550 nm) for the ZrO_2 - VO_2 layer system. The deposition was completed by the top zirconium oxide layer of 170 nm thickness. The layer thickness for all three layers was kept constant. The choice is based on results reported in the literature (Houska et al., 2019). They found that this layer structure provided the highest values for both T_{lum} and ΔT_{sol} .

Samples of the size 200 mm x 200 mm were cut out of the continuous roll for detailed investigation. The transmission and reflection spectra were measured by a spectrophotometer with integrating sphere (Perkin Elmer Lambda 1150).

3 RESULTS

Different thermochromic layer systems were investigated. They differ in both the optical properties and the transition temperature. The deposition method described in the previous section provided stable and reproducible properties of the layers. A comprehensive description of the coating process itself is beyond the scope of this paper. In the following, typical results will be presented, and possible application scenarios of such coatings will be discussed.

Figure 5 shows the transmission spectra of the sample with the highest ΔT_{col}



FIG. 5 Left: Transmission spectra of cold and warm states of a ZrO2-VO2-ZrO2 coating on flexible glass: Right: Thermal hysteresis of the optical properties, plotted for the transmission value at 2500 nm wavelength

The left-hand side of Figure 5 shows the transmission spectra of the sample in the low- and high-temperature modes. The solar spectrum AM1.5 is included in Figure 5 for reference. The spectra of the high- and low-temperature modes are nearly identical in the visible spectral range. However, starting from the wavelength of 1000 nm upwards, the transmittance in the low-temperature mode is higher, reaching a maximum of 65%. The change of the solar energy transmittance ΔT_{sol} between the high- and low-temperature modes amounts to 9.6% for this sample. At the same time, the luminous transmittance T_{lum} of 50% remains unchanged during the phase transition. The right-hand side of Figure 5 shows the hysteresis in the switching behaviour between the high- and low-temperature modes. For reference, the approximate daily variation of the temperature in Athens is given for both summer and winter time. As shown, the sample is subject to seasonal and daily modulation effects under the climatic conditions of Greece. The coating shown in Figure 5 is the basis of all further investigations.

The thermochromic properties of the samples from this study are compared to the state-of-the-art values reported in review articles published in literature. An overview of the achievable properties is given in (Aburas et al., 2019) (Figure 6).



FIG. 6 Typical region for different kinds of vanadium oxide-based thermochromic coatings in the ΔTcol-Tlum plane. The best obtained results reported in this paper are indicated by an orange square, and the blue area represents the expected performance range of the multilayer approach with an antireflection layer. The dashed blue line illustrates the range which can be covered by varying the V02 thickness based on the present level of technology, and circled areas are adopted from (Aburas et al., 2019).

The multilayer approach with the antireflection layer is represented by the blue field in Figure 6. This corresponds to the type of coating investigated in this article. The results show that the performance values achieved on flexible glass are in the same range as those achieved by other research groups on sheet glass. The multilayer approach itself is only outperformed by the nanoparticle approach. A detailed analysis of this type of coating can be found in (Wang, 2021).

Based on the optical measurements, the temperature-dependent refractive index *n* and extinction coefficient *k* were determined. Using these values, ΔT_{sol} and T_{lum} could be simulated (dashed blue line in Figure 6). Provided that the optical constants do not depend on the layer thickness, this gives an insight into potential performance improvements. The curve in Figure 6 indicates that an increase in ΔT_{sol} can be achieved at the expense of the luminous transmittance. This result illustrates the applicability of the technology in scenarios with excessive availability of light.

As it was outlined in the introduction, the potential of smart coatings should be evaluated in combination with state-of-the-art solutions. Therefore, the data given in Figure 5 and Figure 6 were used to calculate the performance in various combinations of window coatings shown in Figure 7.





The effect of the low-e-and solar control coatings is simulated by introducing a typical layer stack $(TiO_2 (30 \text{ nm})-Ag (10 \text{ nm})-TiO_2-(30 \text{ nm}))^n$, adopted from (Solovyev et al., 2015). A low-e coating corresponds to n = 1, and the solar control stack stands for n = 2. The solar control version has a sharper drop in transmittance in the near-infrared spectral range, thus preventing the thermal radiation of the sun from entering the building. Each of these coatings is combined with the thermochromic layer stack on one glass surface.

Figure 8 shows the calculated ΔT_{sol} in these different combinations.



FIG. 8 Change of solar transmittance ΔTsol for a thermochromic coating in different window coating combinations.

It is apparent that the combination of thermochromic coatings with state-of-the-art low-e coatings and solar control coatings does not offer any benefit to the overall optical properties of the window. Here, the change in the optical properties (ΔT_{so}) of the entire window is remarkably suppressed and, in some cases, drops to negligible values of around 1%. The implementation of low-e-coatings is important and inevitable for the efficiency of heating systems in temperate and cold climates. Therefore, thermochromic coatings cannot improve the thermal insulation of such buildings efficiently.

In contrast to these examples, thermochromic coatings can be advantageous if applied in buildings that require permanent cooling. This coincides with findings presented in the literature (Tällberg et al., 2016). During the daytime, they are in a warm state and block solar radiation, similar to low-e coatings. In the nighttime, the thermochromic material switches to the cold state. Then, it outperforms the static low-e and solar control coatings. In its cold state, the thermochromic layer behaves like a non-conductive ceramic. It does not prevent the heat transport from the inside to the outside of the building. In the same way, it supports radiative cooling because the thermal radiation can escape the hot building to the colder outside in the same way as if the window glass of the building was completely uncoated. In contrast to that, state-of-the art low-e coatings drastically reduce the possible energy exchange by thermal radiation.

The radiative heat exchange is especially effective for skylights. The sky is a heat sink for thermal radiation. In the mid-infrared spectral range between 3 μ m and 25 μ m, the optical properties of the sky are mainly determined by the water vapour content of the air. Thus, they are dependent on the local climatic conditions of the installation as well as on the weather. The emissivity of the sky is in the range of 0.9 (Agarni & Nutter, 2015). The effective temperature of the sky can vary considerably, but in most cases, it can be assumed to be at 0°C. In tropical conditions, it can be as low as -50°C (Algarni et al., 2015). If buildings in these areas need skylights for esthetical or other reasons, the application of thermochromic coatings is a viable option for decreasing the energy demand for cooling.



FIG. 9 Picture (left) and schematic design drawing (right) and mockup for the test of thermochromic coatings in transparent roof elements.

The effect of blocking the solar irradiation during the daytime and supporting radiative cooling during the night was tested with a set of mockups as shown in Figure 9.

The design of the mockups is shown on the right side of Figure 9. The mockups are identical, wellisolated boxes with a 22 cm x 30 cm base area and a height of 16 cm. Three identical mockups with different coatings on the transparent cover ("Specimen") were compared with respect to their inside temperature. Each of the mockups was covered with 4 mm float glass. The dimension of the transparent cover was 20 cm x 20 cm. For the first mockup, an ultra-thin glass layer with a thermochromic coating was laminated to the bottom side of the glass. For the second mockup, a PET film with a solar control coating was laminated, replicating the layer stack described in (Solovyev, 2015). For the third mockup, the glass was left uncoated. The properties of the thermochromic coating required a special adaptation to the low-temperature conditions in Dresden, Germany, at the time of the test. They were adapted to both the geographic location and the temperature conditions during the testing (between -2°C and 12°C). The hysteresis of the specially designed coating is shown in Figure 10 (right).





The transition temperature of the coating was determined by the hysteresis data shown in Figure 10 (right). It was calculated according to the integral equation approach (see equation (4)) as $\Theta_{\mu} = 16^{\circ}$ C.

The temperature range during the 24h test is indicated by the crosshatched box in Figure 10 (right). It is slightly below the transition temperature, but the optical properties of the coating show enough modulation within the box.

The diagram on the left-hand side of Figure 10 shows three obtained temperature curves. During the cooling-down phase at night, the temperature in the box with the thermochromic coating followed the reference box with the uncoated glass. The cooling rate for these two mockups was higher than for the mockup with the solar control coating. Under sunshine conditions in the morning, the temperature increase of the mockup with the uncoated glass was excessive. On the other hand, the thermochromic coating as well as the standard static solar control coating significantly damped the increase in temperature.

Overall, the thermochromic coating showed a daily modulation that continuously ensured optimum cooling support for the mockup. This result coincides with the findings reported in literature (Tällberg et al., 2016), (Butt et al., 2021). A deviation from the expected behaviour was observed in the early morning hours around 6 am. An explanation of this phenomenon requires further investigation and longer observation time.

This first test has demonstrated the potential of skylights with thermochromic coatings. It featured results under ideal conditions of a clear sky and low air humidity. For a more profound evaluation, further investigations under various weather conditions over a longer time are necessary. In particular, it is useful to perform the test in a climatic area that primarily requires cooling buildings rather than heating. It is noteworthy how solar modulation, normally referred to as the crucial parameter for the evaluation of the thermochromic effect, loses its importance in the roof application scenario. The support of radiative cooling can be described more precisely by the relationship between daytime solar transmittance and nighttime emissivity. A deeper investigation of this interdependence will be the focus of further scientific work on thermochromic coatings.

4 CONCLUSIONS

VO₂ thermochromic coatings were sputter-deposited on ultra-thin glass in a roll-to-roll coating machine. The width of the continuous roll was 300 mm, and the areal weight of the thermochromic material was lower than 0.3 kg/m². This makes it an attractive option for building retrofit. A modulation of the solar transmission between the high and low-temperature states of 9.6% was achieved. The luminous transmittance amounted to 50% in both states. The transition temperature of 22°C was in the comfortable ambient temperature range. These values correspond well with results reported for deposition processes on sheet glass. Various application scenarios were investigated. The combination with state-of-the-art low-e or solar control coatings drastically reduced the modulation of the solar transmittance. Therefore, thermochromic coatings were shown to be ineffective for heat insulation purposes in cold and temperate climates. For an alternative use case, an implementation in transparent roof elements of buildings which need permanent cooling was tested. Such a configuration reduces the solar transmittance during the daytime and supports radiative cooling during the night.

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