

Broadband-modulated Photochromic Smart Windows Based on Oxygen-containing Gadolinium Hydride Films

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Abstract: Photochromic windows are considered an effective and energy-efficient smart window due to their simple structure, passive light modulation, and zero-energy input. However, the research on photochromic smart windows has rarely addressed the mid-infrared (MIR) bands, which greatly limits energy-saving efficiency. Here, we report that rare-earth oxygen-containing hydrides (ReO_xH_y) films grown on ITO substrates have the ability to be broadband modulated. The developed photochromic smart window is capable of automatically adjusting emissivity by sensing light intensity while maintaining visible and near-infrared (NIR) modulation ($\Delta T_{\text{sol}} = 35.1\%$, $\Delta T_{\text{lum}} = 37\%$, and $\Delta \varepsilon_{8-13\mu\text{m}} = 0.12$). We also achieved one-step preparation of GdO_xH_y with improved stability by optimising the preparation atmosphere. The photochromic mechanism was analyzed by comprehensive characterization. In conclusion, this passive and synergistic modulation method across the visible-NIR-MIR is expected to greatly advance the field of photochromic smart windows.

Key words: photochromic; rare-earth oxygen-containing hydride; smart window; dynamic radiative cooling

Buildings account for approximately 40% of global energy consumption in which their windows, as the primary pathway for energy loss, contributing up to 60%^[1-3]. Fortunately, smart windows can control the amount of heat escaping or injecting into an enclosed space by regulating the transmittance and reflectance of solar radiation spectra^[4-5]. The above helps to reduce the need for indoor temperature regulation in order to save energy. Adaptive smart windows are considered cost-effective because of their simple structure, passive light modulation, and zero-energy input property^[6-7]. Among them, photochromic smart windows stand out for their high optical contrast and adaptive daylight management property^[8].

Additionally, mid-infrared (MIR) atmospheric transparent window helps to exchange thermal radiation between the terrestrial surface and the 3 K outer space, which is a passive way to cool buildings^[9]. High MIR emissivity of smart windows allows for more efficient

cooling. However, fixed high emissivity can lead to overcooling due to loss of indoor heat in cold environment, thus necessitating the development of dynamic radiative cooling (DRC) methods^[10-11]. An ideal photochromic smart window should achieve DRC in addition to solar spectrum modulation. Unfortunately, research on photochromic smart window has so far been predominantly confined to the visible and near-infrared (NIR) regions, neglecting the study of MIR thermal radiation^[12].

As a novel photochromic material, rare-earth oxygen-containing hydride (ReO_xH_y) photochromic films were firstly discovered in 2011 and exhibited highly efficient and reversible photo-darkening property^[13-14]. ReO_xH_y demonstrates substantial modulation across visible and NIR wavelength bands, surpassing other inorganic photochromic materials in modulation range^[15-16]. Nonetheless, achieving controlled preparation remains a daunting task owing to the significant impact of oxygen

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content on performance^[17]. Despite its excellent broadband modulation potential, it has been neglected by researchers.

In this study, we present a broadband-modulated photochromic smart window consisting of an ITO/GdO_xH_y bilayer film. This film exhibits notable spectral modulation from the visible to NIR before and after photoexcitation, with $\Delta T_{\text{sol}} = 35.1\%$ and $\Delta T_{\text{lum}} = 37\%$. Most importantly, it extends modulation capability into the MIR region ($\Delta \varepsilon_{8-13 \mu\text{m}} = 0.12$). The fabrication process is simple and cost-effective, involving only magnetron sputtering at room temperature. Additionally, we delve into the optimal preparation conditions, the photochromic mechanism, and bleaching rate of GdO_xH_y films, laying the foundation for this photochromic smart windows in this innovative system.

1 Experimental

1.1 Materials and synthesis

The GdO_xH_y thin films were deposited using a ULVAC-ASC-400C4 magnetron sputtering apparatus with a 100 W direct current (DC) power. A two-inch metallic Gd target with 99.9% purity was used as the sputtering target. The sputtering gases were hydrogen and argon, both with a purity of 99.99%, and their flow rates were measured in sccm (Standard Cubic Centimeters per Minute). The ratio of argon to hydrogen was adjusted to prepare different samples during the experiment. Quartz glass and an ITO substrate were used to deposit the GdO_xH_y films. The sputtering chamber had a background pressure of 5×10^{-4} Pa. The substrate temperature was consistently maintained at room temperature.

1.2 Characterizations

Transmittance was measured from 350 to 2500 nm using a UV spectrophotometer (U-4100, Hitachi High-Technologies). Illumination was carried out using a Xenon lamp (PLS-SXE 300 (UV), PerfectLight, 200 W) or a UV lamp (365 nm, 80 W). The structural phases of the GdO_xH_y films deposited on the substrate were determined using X-ray diffraction (XRD). The analysis was conducted on a Rigaku Ultima IV diffractometer using Cu K α radiation ($\lambda = 0.15418$ nm) in 2θ scanning mode. The morphology and thickness of the films were measured using scanning electron microscope (SEM, Hitachi SU8220) and atomic force microscope (AFM, SII Nanotech Ltd, Nanonavi P). X-ray photoelectron spectroscopy (XPS, ESCA Lab 250, Thermo Fisher Scientific Co. Inc.) was employed to analyze the chemical state of the films. All binding energies were referenced to the C1s peak of carbon (284.8 eV). Infrared

spectra (2.5–25 μm) were measured using a Fourier transform infrared (FT-IR) spectrometer (Thermo Scientific NICOLET Is10). Infrared photographs were taken with an infrared camera (InfRec R300SR) positioned, 50 cm away from the sample and placed at an angle of $\sim 30^\circ$ above the sample.

In order to evaluate the visible transmittance and solar modulation properties of the films, the visible transmittance (T_{lum}) and solar transmittance (T_{sol}) were calculated according to the following equations:

$$T_{\text{lum}} = \frac{\int \Phi_{\text{lum}}(\lambda) T(\lambda) d(\lambda)}{\int \Phi_{\text{lum}}(\lambda) d(\lambda)} \quad (1)$$

$$T_{\text{sol}} = \frac{\int \Phi_{\text{sol}}(\lambda) T(\lambda) d(\lambda)}{\int \Phi_{\text{sol}}(\lambda) d(\lambda)} \quad (2)$$

where $T(\lambda)$ denotes the transmittance at wavelength λ and $\Phi_{\text{lum}}(\lambda)$ is the spectral sensitivity of the light-adapted eye. Φ_{sol} is the spectrum of solar radiation at an air mass of 1.5, which corresponds to solar irradiation at 37° above the horizon. The solar modulation efficiency (ΔT_{sol}) refers to the difference of T_{sol} before and after illumination. Based on the above results, the ΔT_{sol} can be obtained. For the sample prepared in different environments, the maximum change in transmittance was taken as an indicator of the change in performance.

According to Kirchhoff's law, the absorptivity of an object is equal to the emissivity. For non-transparent samples, the emissivity is equal to one minus the reflectivity. Therefore, the infrared emissivity can be calculated from the following formulas:

$$B(\lambda, T) = \frac{2\pi h c_1^2}{\lambda^5} \frac{1}{e^{\left(\frac{hc_2}{\lambda \kappa_B T}\right)} - 1} \quad (3)$$

$$\varepsilon(\lambda) = \frac{\int_{\lambda_{\min}}^{\lambda_{\max}} (1 - R(\lambda)) \cdot B(\lambda, T) d\lambda}{\int_{\lambda_{\min}}^{\lambda_{\max}} B(\lambda, T) d\lambda} \quad (4)$$

where $B(\lambda, T)$ is the spectral emission of the blackbody, κ_B is the Boltzmann constant, h is Planck's constant, c is the speed of light, and λ is the wavelength, and $\varepsilon(\lambda)$ is the spectral emissivity. The difference in optical properties is the difference before and after light irradiation.

2 Results and discussion

2.1 Characterization of GdO_xH_y film

Fig. 1(a) shows the XRD patterns of GdO_xH_y grown on ITO. GdO_xH_y exhibits good crystallinity, and its diffraction peaks can be attributed to the crystallographic bixbyite Gd₂O₃ (space group Ia $\bar{3}$) and fcc GdH₂ (space

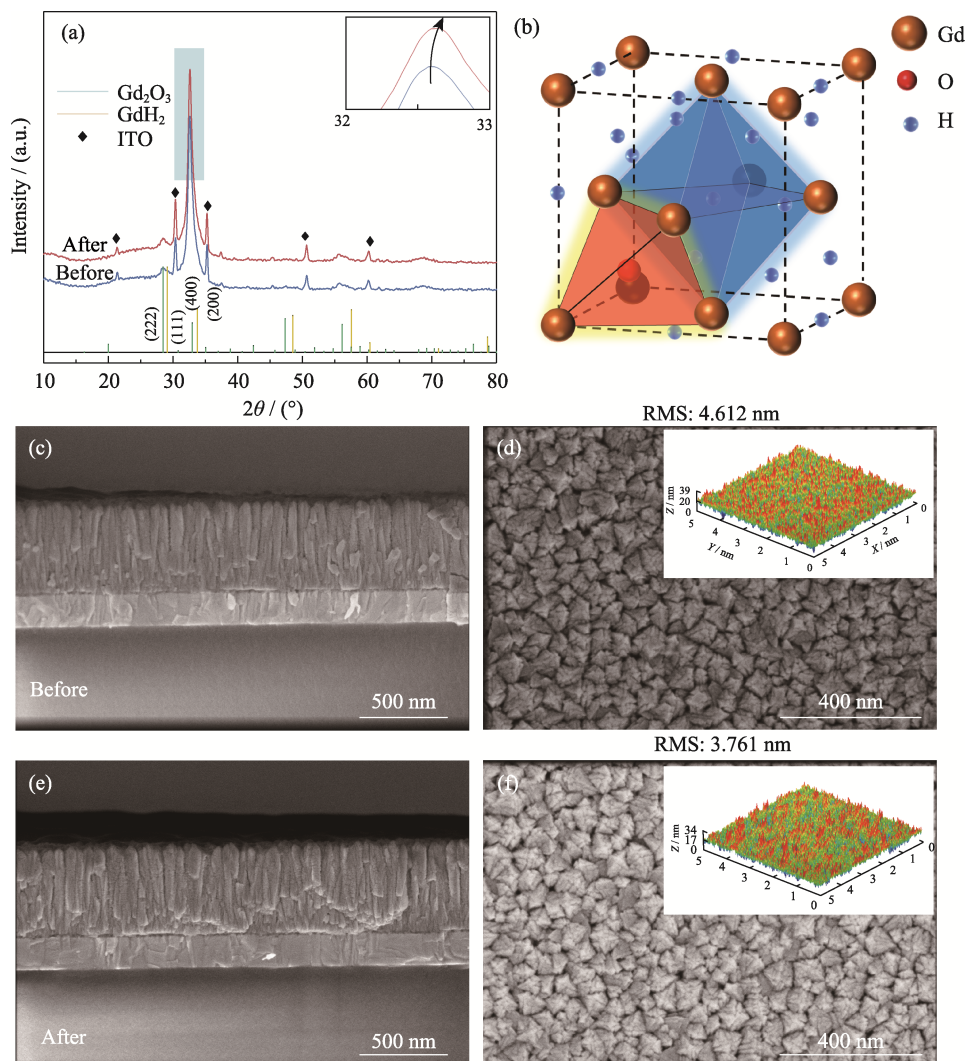


Fig. 1 Microstructural characterization of ITO/GdO_xH_y bilayer film

(a) XRD patterns of ITO/GdO_xH_y before (blue) and after (red) illumination; (b) Schematic crystal structure of GdO_xH_y;
(c-f) SEM images of cross section (c, e) and surface (d, f) of thin film samples with the insets in (d, f) showing the corresponding AFM images

group $Fm\bar{3}m$) phases. The absence of further peaks after illumination suggests that the films are composed of the same crystalline substance. After illumination, the diffraction peaks of the GdO_xH_y films exhibit a slight shift by a significant angle. This is in line with previous findings showing that the GdO_xH_y films underwent lattice shrinkage^[18]. This contraction may result from the release of oxygen from the lattice. The lattice contraction under light can also be explained by the light-induced proton injection. As the concentration of H increases, the octahedral sites become filled, leading to lattice compression (Fig. 1(b))^[19].

Fig. 1(c-f) show the cross-section and surface SEM images of the ITO/GdO_xH_y films, along with the corresponding AFM results. Consistent with previously reported rare-earth oxyhydroxides exhibiting photodarkening properties, the cross-section of the GdO_xH_y film shows a continuous columnar growth pattern. The surface displays a denser, needle-like crystal morphology,

which is associated with the penetration of oxygen into the film. This structure may generate residual stress within the film, providing a driving force for proton migration^[20]. The film layer demonstrates good flatness both before and after illumination. The AFM results indicate that the average roughness of the film layer is less than 5 nm for thicknesses exceeding 700 nm.

We investigated the compositional changes on the surface of GdO_xH_y films using XPS (Fig. 2). The signal related to C–C is set to 284.80 eV as a charge correction reference. A second contribution is observed at 289.05 eV, which is attributed to O–C=O^[21]. In the C–C peak position, the carbon content on the surface decreases after exposure to light. The reduction in the adsorption of carbon-containing impurities may be caused by atomic rearrangements on the surface^[22]. The O1s signal consists of two components at 529.19 and 531.19 eV. The former can be attributed to O atoms binding to Gd atoms, while the latter corresponds to signals from

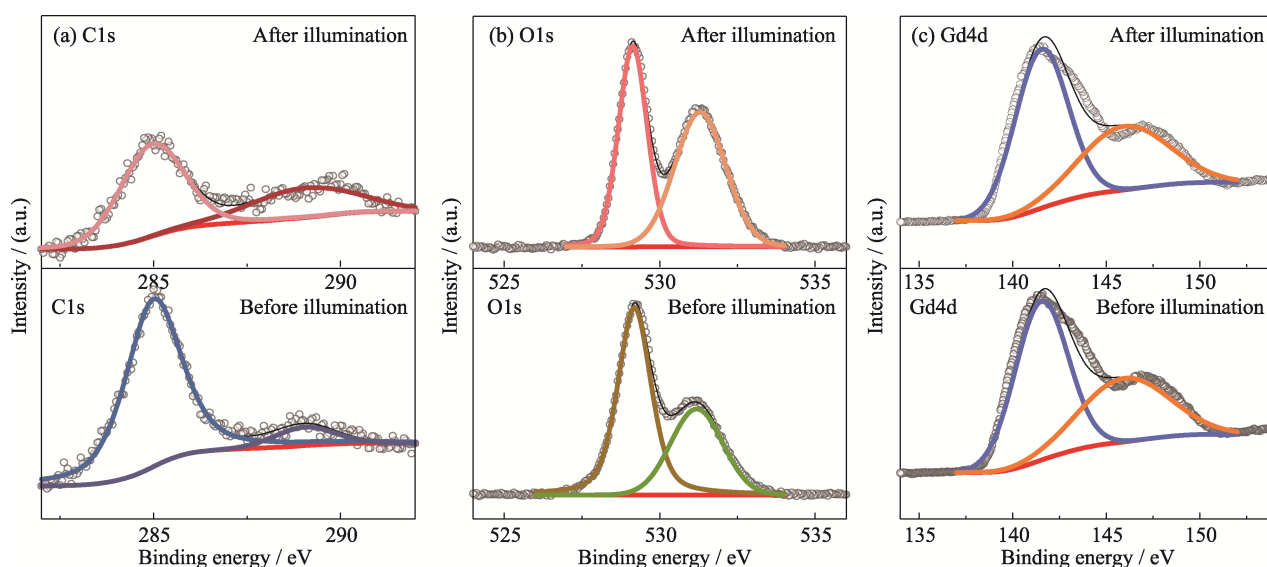


Fig. 2 XPS spectra of ITO/GdO_xH_y before and after the illumination
(a) C1s; (b) O1s; (c) Gd4d

atomic oxygen. The ratio of O to Gd atoms on the surface increase after exposure to light. This enhancement indicates oxygen enrichment at the surface^[23]. The experimental results for Gd4d reveal concentrated characteristic peaks at 141.50 and 145.98 eV. However, the data fitting is not optimal, mainly due to the Gd4d spectra exhibit nuclear level splitting caused by spin-orbit coupling in the Gd4d spectra. The strongest spectral feature results from the interaction of the 4d electrons with the valence band of 4f^[24]. Gd4d does not show significant chemical energy shift, and the content does not undergo significant difference. Based on the above characterization, we believe that this broadband response property originates from the rearrangement of hydrogen and oxygen in the vertical direction of the film after illumination. This is manifested by the enrichment of oxygen near the surface of the film and the formation of a localized GdH₂ phase in the interior^[17,25].

2.2 Stability and bleaching rate of GdO_xH_y films

Smart windows utilizing ReO_xH_y film have made significant progress toward practical applications, but two key issues remain unresolved: stability and bleaching rate^[26]. ReO_xH_y films contain non-stoichiometric oxygen and hydrogen ions. Research has demonstrated that the film's composition has a dramatic impact on its photochromic properties^[27]. The current main growth preparation method involves firstly forming a hydrogenated phase with a low oxygen content, followed by a slow oxidation process in air to obtain an optimal ReO_xH_y phase. But oxidation takes a lot of time, usually one day to one week. This makes it hard to use the sample in real life. In response to this challenge, recent

studies have explored new approaches to achieve optimal performance of photochromic smart windows. The direct preparation of GdO_xH_y by manipulating the gas flow rate under relatively low vacuum conditions, leading to promising results^[28].

In this study, we explore the optimal atmosphere for growing thin films using a one-step method in order to achieve stable broadband modulation performance. The XRD analysis in Fig. 3(a) shows that as the argon to hydrogen ratio of the sputtering atmosphere increases from 40:10 to 40:30, the characteristic peaks of the films gradually weaken and shift (growth time was 30 min, quartz glass substrates). The variance in transmittance in the visible region for the different samples is summarized in Fig. 3(b). At lower sputtering pressures (25:15 and 30:10), the films do not exhibit photochromic properties. Instead, the films with high hydrogen content appear opaque black and require a post-oxidation process to achieve color-changing capability. When the Ar:H₂ ratio is 40:10, transparent GdO_xH_y films have a maximum transmittance exceeding 80% and a transmittance difference of 45%. However, further increasing the flow rate of hydrogen leads to a decrease in film properties and initial transmittance. To assess the bleaching rate, films prepared at 40:10 and 40:20 were tested, and the results are presented in Fig. 3(c). After a sufficiently long bleaching period in a dark environment, both films can regain over 80% transmittance. This observation demonstrates the favorable bleaching rate at which the one-step prepared films.

The stability tests were conducted simultaneously for the 40:10 and 40:20 films in a constant temperature and humidity chamber (at room temperature, 60% relative humidity). Fig. 3(d, e) illustrate the transmittance

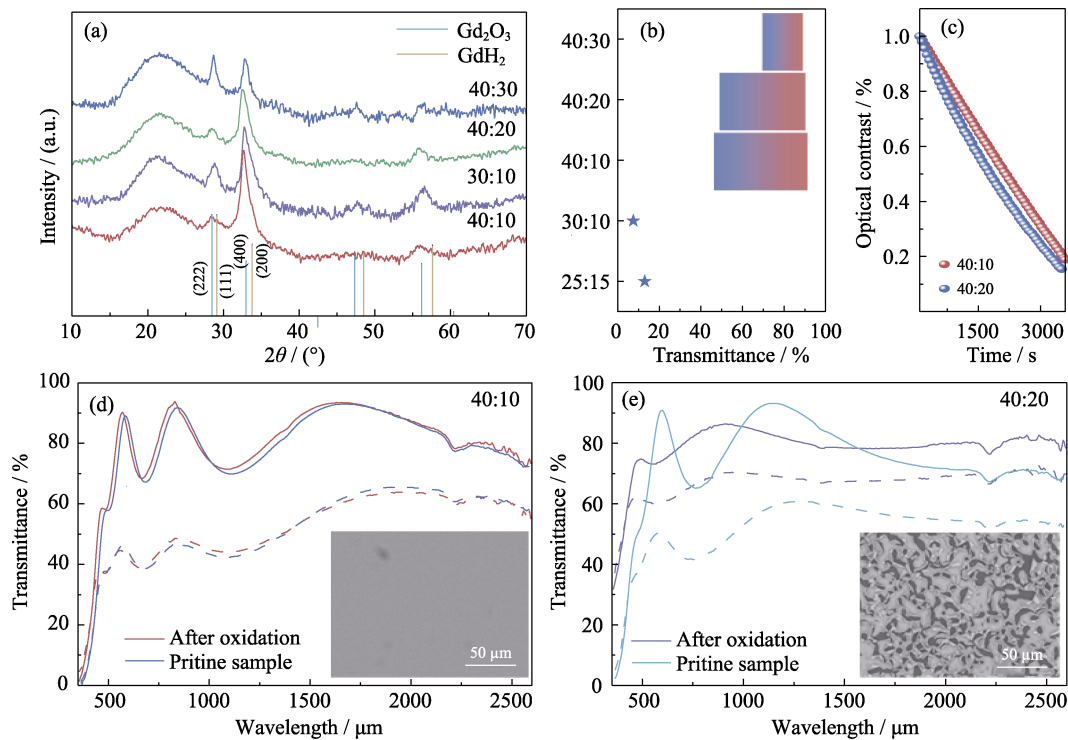


Fig. 3 Effect of sputtering atmosphere on film properties

(a) XRD patterns of films prepared under different sputtering atmospheres; (b) Value at the point of maximum change in transmittance for films grown under different sputtering atmospheres; (c) Bleaching time of films under different sputtering atmospheres; (d, e) Transmittance spectra of films before and after stability test

spectra of the respective samples, with the inset displaying the SEM images of the surface after 10 d of placement. The 40:20 films exhibit degradation of properties. Conversely, the transmittance spectra for the 40:10 sample reveal that 10 d of natural oxidation does not compromise the photochromic properties, and the surface morphology remains relatively intact. The difference between the films is attributed to the variance in hydrogen content, with negatively valenced hydrogen ions being more susceptible to reacting with oxygen in the air. The films can achieve optimal sputtering efficiency, attenuation rate, and stability when the hydrogen-to-oxygen ratio is balanced.

The films were subjected to different temperature fields in order to determine the rate of discolouration. The coloring process, illustrated in Fig. 4(a), is facilitated by the excitation of photon as it crosses the energy barrier. However, the bleaching speed is relatively slow due to the absence of external energy intervention. Applying a thermal field accelerates the fading process and effectively reduces the energy potential barrier of the bleaching process (Fig. 4(b)). As shown in Fig. 4(c), there is almost no colour fading after 20 min at sub-zero temperature. When the films are placed at room temperature, the optical contrast ΔT recovers by about 30% after 20 min of fading process. The temperature is raised to 50 °C, and after 20 min, ΔT recovers by 40%.

Furthermore, at 70 °C, about 80% of the starting state can be recovered in 10 min, and almost complete recovery can be achieved within 20 min (the corresponding transmission spectrum is shown in Fig. 4(d)). However, excessively high temperature can alter the film components and subsequently impact the photochromic properties. Before being exposed to light, uniformly prepared films were subjected to atmospheric pressure annealing at 100, 150, and 200 °C, and compared with films without annealing treatment. A comparison of the photochromic properties is shown in Fig. 4(e, f), revealing a gradual decrease with increasing annealing temperature. At temperature above 100 °C, the photochromic performance decreases because the annealed materials absorb less light. This happens because there is more oxygen in the material during high-temperature oxidation. This is indicated by the increasing optical band gap, which rises from 2.06 to 2.26 eV, resulting in a decrease in photochromic properties^[29].

2.3 Broadband photochromic smart windows

Fig. 5 shows the spectra of the ITO/GdO_xH_y film. In previous studies, GdO_xH_y has been found to potentially involve the migration of hydrogen atoms and the detachment/trapping of oxygen atoms before and after discoloration^[30-31]. The ITO substrate can reflect the infrared, and provide transparency in the UV-visible

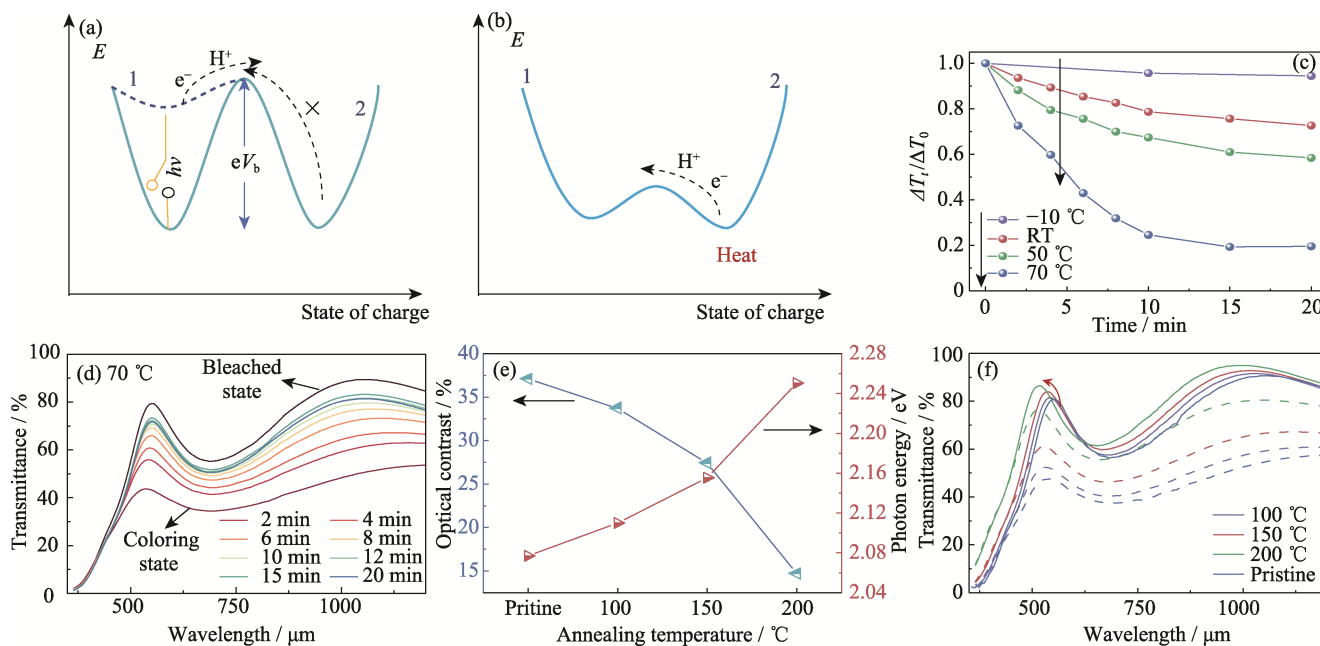
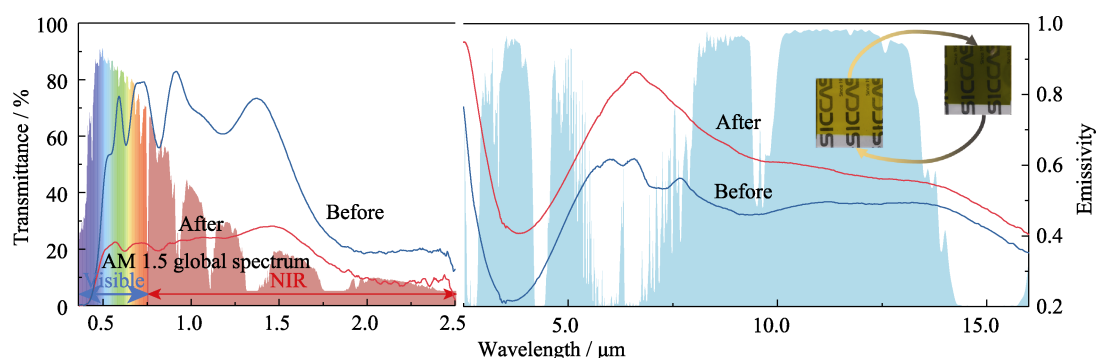


Fig. 4 Effect of temperature on the bleaching rate

(a) Energy-barrier relationship between the "bleached state (1)" and "coloring state (2)" at room temperature; (b) Energy-barrier relationship between "bleached state (1)" and "coloring state (2)" when heated; (c) Bleaching rates of films at different temperatures; (d) Variation of transmission spectra of films with time and placed in a temperature field of 70 °C; (e) Relationship between optical contrast, band gap (Tauc-plot method), and annealing temperature; (f) Transmittance spectra of GdO_xH_y films at different annealing temperatures; Colorful figures are available on website

Fig. 5 Transmittance and emissivity spectra of the ITO/GdO_xH_y bilayer film before and after light illumination

region to allow for further tuning. The spectrum shows that the transmittance of this sample in the visible-NIR band decreases dramatically, leading to substantial solar modulation efficiencies ($\Delta T_{\text{sol}} = 35.1\%$, $\Delta T_{\text{lum}} = 37\%$). More importantly, its emissivity increases in the atmospheric window band after exposure to light ($\Delta \varepsilon_{8-13 \mu\text{m}} = 0.12$).

An infrared camera was used to detect how the emissivity of the films changed over time. The films were placed in a UV light field with an ITO substrate and quartz glass, as depicted in Fig. 6(a). As the illumination continued, the ambient temperature gradually increased, but the temperature distribution across the films was uniform. The gradual increase in emissivity observed on the surface of the ITO/GdO_xH_y films led to a more pronounced change in surface emission temperature

compared to the low emissivity surface (ITO) and the high emissivity surface (glass).

The sample surface can be pattern etched as shown in Fig. 6(b). This etching process was done using a mask projection technique in which a mask plate was used to block specific areas of the sample and let UV light shine on them. The nature of the photo response makes it easy to manipulate, so we can pattern films with light projection and UV direct writing approaches for information and energy management. Capturing images of the sample at visible and infrared wavelengths (8–14 μm) using a camera with a specific wavelength provide clear and unambiguous patterning information. The emissivity of the sample in the illuminated region is notably increased, making it easily distinguishable from the background.

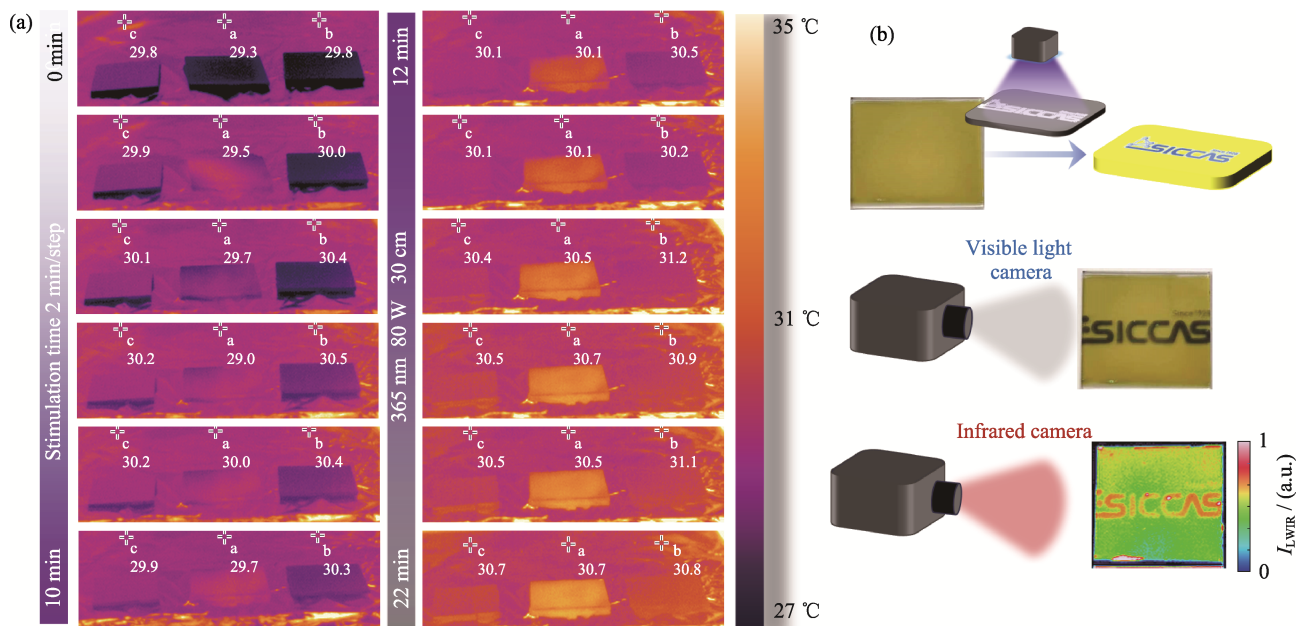


Fig. 6 Demonstration of film versatility

- (a) Variation of emission temperature of ITO (left), ITO/GdO_xH_y (middle), and glass (right) under UV irradiation;
 (b) Optical lithography of the "SICCAS" pattern on the surface of the sample by UV light projection

3 Conclusions

In this study, we present a pioneering approach to create a photochromic smart window capable of broad spectral modulation. This method leverages the unique optical properties of rare-earth oxygenated hydrides to achieve significant modulation, with $\Delta T_{\text{sol}} = 35.1\%$, $\Delta T_{\text{lum}} = 37\%$, and $\Delta \varepsilon_{8-13 \mu\text{m}} = 0.12$. Importantly, our design is straightforward, free from intricate electronic circuits, facilitating easy fabrication and implementation. Furthermore, when combined with UV projection technology, these smart windows can be conveniently patterned on surfaces. Notably, the implications of this work extend beyond simple window technology, offering the potential for the development of dynamic devices with a broader application scope, including infrared data storage, encryption, camouflage, and thermal management.

References:

- [1] FUMO N. A review on the basics of building energy estimation. *Renewable & Sustainable Energy Reviews*, 2014, **31**: 53.
- [2] MA L D, SHAO N N, ZHANG J L, *et al.* The Influence of Doors and Windows on the Indoor Temperature in Rural House. 9th International Symposium on Heating Ventilation and Air Conditioning ISHVAC Joint with the 3rd International Conference on Building Energy and Environment COBEE, Tianjin, 2015: 621–627.
- [3] XU F, JIN P S, LUO H J, *et al.* VO₂ thermochromic smart window: status, challenges and prospects. *Journal of Inorganic Materials*, 2021, **36**(10): 1013.
- [4] ZAKIRULLIN R S. Chromogenic materials in smart windows for angular-selective filtering of solar radiation. *Materials Today Energy*, 2020, **17**: 9.
- [5] NGUYEN T D, YEO L P, ONG A J, *et al.* Electrochromic smart glass coating on functional nano-frameworks for effective building energy conservation. *Materials Today Energy*, 2020, **18**: 10.
- [6] WANG S F, LIU M S, KONG L B, *et al.* Recent progress in VO₂ smart coatings: strategies to improve the thermochromic properties. *Progress in Materials Science*, 2016, **81**: 1.
- [7] MENG W, KRAGT A J J, GAO Y, *et al.* Scalable photochromic film for solar heat and daylight management. *Advanced Materials*, 2024, **36**(5): 2304910.
- [8] SHAN Z W, CAO X, ZHANG Q X, *et al.* Fully automatic modulation for broadband sunlight based on YO₃H₂/VO₂ hybrid structures. *Solar Energy Materials and Solar Cells*, 2019, **200**: 9.
- [9] DONG M H, CHEN N, ZHAO X D, *et al.* Nighttime radiative cooling in hot and humid climates. *Optics Express*, 2019, **27**(22): 31587.
- [10] WANG S C, JIANG T Y, MENG Y, *et al.* Scalable thermochromic smart windows with passive radiative cooling regulation. *Science*, 2021, **374**(6574): 1501.
- [11] ZHANG Q, WANG S H, WANG X Y, *et al.* Recent progress in daytime radiative cooling: advanced material designs and applications. *Small Methods*, 2022, **6**(4): 19.
- [12] WANG S F, FAN W R, LIU Z C, *et al.* Advances on tungsten oxide based photochromic materials: strategies to improve their photochromic properties. *Journal of Materials Chemistry C*, 2018, **6**(2): 191.
- [13] CORNELIUS S, COLOMBI G, NAFEZAREFI F, *et al.* Oxyhydride nature of rare-earth-based photochromic thin films. *Journal of Physical Chemistry Letters*, 2019, **10**(6): 1342.
- [14] LI M, SHAO Z W, LI Z S, *et al.* Co-sputtering crystal lattice selection for rare earth metal-based multi cation and mixed anion photochromic films. *Nanomaterials*, 2023, **13**(4): 11.
- [15] ZHANG Q X, XIE L L, ZHU Y, *et al.* Photo-thermochromic properties of oxygen-containing yttrium hydride and tungsten oxide composite films. *Solar Energy Materials and Solar Cells*, 2019, **200**: 8.
- [16] STRUGOVSHCHIKOV E, PISHTSHEV A, KARAZHANOV S. Theoretical design of effective multilayer optical coatings using oxyhydride thin films. *Physica Status Solidi B-Basic Solid State Physics*, 2021, **258**(10): 7.
- [17] MOLDAREV D, STOLZ L, MORO M V, *et al.* Environmental dependence of the photochromic effect of oxygen-containing rare-earth metal hydrides. *Journal of Applied Physics*, 2021, **129**(15): 7.
- [18] HANS M, TRAN T T, ADALSTEINSSON S M, *et al.* Photochromic mechanism and dual-phase formation in oxygen-containing rare-

- earth hydride thin films. *Advanced Optical Materials*, 2020, **8**(19): 6.
- [19] NAFEZAREFI F, CORNELIUS S, NIJSKENS J, *et al.* Effect of the addition of zirconium on the photochromic properties of yttrium oxy-hydride. *Solar Energy Materials and Solar Cells*, 2019, **200**: 6.
- [20] PETROV I, BARNA P B, HULTMAN L, *et al.* Microstructural evolution during film growth. *Journal of Vacuum Science & Technology A*, 2003, **21**(5): S117.
- [21] KHAN S, AZIMI G, YILDIZ B, *et al.* Role of surface oxygen-to-metal ratio on the wettability of rare-earth oxides. *Applied Physics Letters*, 2015, **106**(6): 5.
- [22] ULLAH N, IMRAN M, LIANG K, *et al.* Highly dispersed ultra-small Pd nanoparticles on gadolinium hydroxide nanorods for efficient hydrogenation reactions. *Nanoscale*, 2017, **9**(36): 13800.
- [23] KULAH E, MAROT L, STEINER R, *et al.* Surface chemistry of rare-earth oxide surfaces at ambient conditions: reactions with water and hydrocarbons. *Scientific Reports*, 2017, **7**: 10.
- [24] ZAVABETI A, OU J Z, CAREY B J, *et al.* A liquid metal reaction environment for the room-temperature synthesis of atomically thin metal oxides. *Science*, 2017, **358**(6361): 332.
- [25] YOU C C, KARAZHANOV S Z. Effect of temperature and illumination conditions on the photochromic performance of yttrium oxyhydride thin films. *Journal of Applied Physics*, 2020, **128**(1): 7.
- [26] MOLDAREV D, MORO M V, YOU C C, *et al.* Yttrium oxyhydrides for photochromic applications: correlating composition and optical response. *Physical Review Materials*, 2018, **2**(11): 6.
- [27] YOU C C, MOLDAREV D, MONGSTAD T, *et al.* Enhanced photochromic response in oxygen-containing yttrium hydride thin films transformed by an oxidation process. *Solar Energy Materials and Solar Cells*, 2017, **166**: 185.
- [28] SHAN Z, CAO X, ZHANG Q, *et al.* Fully automatic modulation for broadband sunlight based on $\text{YO}_x\text{H}_y/\text{VO}_2$ hybrid structures. *Solar Energy Materials and Solar Cells*, 2019, **200**: 110044.
- [29] MAKULA P, PACIA M, MACYK W. How to correctly determine the band gap energy of modified semiconductor photocatalysts based on UV-Vis spectra. *Journal of Physical Chemistry Letters*, 2018, **9**(23): 6814.
- [30] BABA E M, MONTERO J, STRUGOVSHCHIKOV E, *et al.* Light-induced breathing in photochromic yttrium oxyhydrides. *Physical Review Materials*, 2020, **4**(2): 8.
- [31] MAEHLEN J P, MONGSTAD T T, YOU C C, *et al.* Lattice contraction in photochromic yttrium hydride. *Journal of Alloys and Compounds*, 2013, **580**: S119.

基于含氧氢化钇薄膜的宽波段调制光致变色智能窗

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摘 要: 光致变色窗户因结构简单、被动调光和零能耗输入而被认为是一种高效的节能智能窗, 然而有关光致变色智能窗的研究很少涉及中红外(MIR)波段, 这限制了节能效率。本研究通过在 ITO 衬底上生长稀土含氧氢化物(ReO_xH_y)薄膜, 实现了宽波段调制的能力。所开发的光致变色智能窗口能够通过感应光强度自动调节发射率, 同时保持可见光和近红外(NIR)调制($\Delta T_{\text{sol}}=35.1\%$, $\Delta T_{\text{lum}}=37\%$, $\Delta \varepsilon_{8-13\text{ }\mu\text{m}}=0.12$)。研究还通过优化制备气氛, 实现了 GdO_xH_y 的一步制备并提高了稳定性, 通过综合表征分析了光致变色机理。总之, 这种跨越可见光-近红外-中红外的被动协同调制方法有望推动光致变色智能窗的发展。

关 键 词: 光致变色; 稀土含氧氢化物; 智能窗; 动态辐射制冷

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