



Thermochromic Materials for Smart Windows: A State-of-Art Review

Xuanjie Wang and Shankar Narayan*

Department of Mechanical, Aerospace and Nuclear Engineering, Rensselaer Polytechnic Institute, Troy, NY, United States

Smart windows that regulate solar energy by changing optical characteristics have recently gained tremendous interest for energy-saving and indoor-comfort applications. Among them, thermochromic smart windows are promising because of their simplicity for industrial production and ease of implementation. Although significant advancements have been reported on thermochromic materials, both optical and transition properties remain unsatisfactory. This review focuses on the recent advancement of thermochromic materials for smart windows in terms of operation, performance, and potential for commercialization. It discusses the parameters typically used for gauging performance and provides a summary and comparison of various promising thermochromic materials, including vanadium dioxide, hydrogels, and perovskites. The article also points the challenges in the practical implementation of these materials and provides an outlook for future development.

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*Correspondence:

Shankar Narayan
narays5@rpi.edu

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INTRODUCTION

With population growth and climate change, the rise in energy consumption has become a critical global issue. A significant portion of the energy consumption in buildings is due to heating, ventilation, and air conditioning (HVAC). Numerous studies have focused on materials with tunable optical properties to regulate and convert solar energy for different applications (Li et al., 2020a, Li et al., 2020b; Wang et al., 2020a; Wang et al., 2020b; Wang et al., 2020c). Since windows are substantial sources of heat to the building envelopes, recent investigations on smart windows aim to reduce energy consumption while maintaining thermal comfort for the indoor environment (Khandelwal et al., 2017; Rezaei et al., 2017).

Smart windows are glazed units coated with unique materials to have dynamic control of the transmittance of solar irradiation into buildings under different ambient environments. Typically, based on the glazing characteristics and operation, smart windows can be categorized under actively dynamic or passively dynamic systems. Active dynamic systems change the optical characteristics with an external stimulus, including electricity, mechanical strain, and chemicals (Baetens et al., 2010; Casini, 2018). Passive dynamic systems, on the contrary, respond to a natural stimulus without the input of an artificial external stimulus. Due to the automatic tunability of optical properties without active intervention, smart windows with passive dynamic systems are gaining traction in various applications.

Passive dynamic smart windows can be further categorized according to the respective natural stimulus, including heat, light, and humidity, resulting in thermochromic, photochromic, and humidity-chromic systems. For photochromic windows, a change in the color of constituent materials, including organic and hybrid organic-inorganic dyes, is activated by sunlight (Wu et al., 2017). On the other hand, humidity-chromic windows are activated by changes in

ambient humidity resulting in the variation of optical properties (Nandakumar et al., 2018). A temperature variation in the environment naturally triggers the optical properties of thermochromic devices. Lately, significant advancements have been reported on thermochromic smart windows due to their operational simplicity, scalable production, and ease of implementation (Ke et al., 2018; Aburas et al., 2019). While previous reviews on thermochromic thin films provide an insightful discussion on different coating technologies and how they compare with commercial smart windows (Kamalisarvestani et al., 2013; Tällberg et al., 2019), this article focuses mainly on recent advances in thermochromic materials, such as vanadium dioxide (VO₂), hydrogels, and perovskites.

The optical and transition properties of thermochromic materials are critical to achieving energy savings in large-scale applications. The crucial properties include the transition temperature (T_c), luminous transmittance (T_{lum}), solar modulation ability (ΔT_{sol}), color, and durability. The luminous transmittance, T_{lum} and solar transmittance, T_{sol} are calculated as shown below (Cui et al., 2018).

$$T_{lum} = \frac{\int_{\lambda=380\text{ nm}}^{780\text{ nm}} T(\lambda)V(\lambda)d\lambda}{\int_{\lambda=380\text{ nm}}^{780\text{ nm}} V(\lambda)d\lambda} \quad (1)$$

$$T_{sol} = \frac{\int_{\lambda=300\text{ nm}}^{2500\text{ nm}} T(\lambda)I_S(\lambda)d\lambda}{\int_{\lambda=300\text{ nm}}^{2500\text{ nm}} I_S(\lambda)d\lambda} \quad (2)$$

The solar modulation ability ΔT_{sol} could be calculated as

$$\Delta T_{sol} = T_{sol}^{cold} - T_{sol}^{hot} \quad (3)$$

Here λ is the wavelength, $T(\lambda)$ is the spectral transmittance of the material, $V(\lambda)$ is the photopic luminous efficiency of the human eye, $I_S(\lambda)$ is the spectral irradiance of the incoming solar radiation, T_{sol}^{cold} and T_{sol}^{hot} are solar transmittance at cold and hot temperatures, respectively (Cui et al., 2018).

The following sections summarize and compare the state-of-art thermochromic smart window materials in terms of operation, performance, and future potential. We focus on three groups of thermochromic materials, which include VO₂, hydrogels, and perovskites. The fundamental mechanisms and recent performance improvement strategies for thermochromic smart windows are also summarized, along with a future outlook for further development.

VANADIUM DIOXIDE

Vanadium dioxide (VO₂) has been widely investigated as a thermochromic material for smart windows due to the reversible metal-to-insulator transition (MIT) at the critical temperature (T_c) of 68°C (Goodenough, 1971). At lower temperatures, the monoclinic (M) insulating phase VO₂ is infrared-transparent, while at higher temperatures, the rutile (R) metallic VO₂ phase is infrared-translucent. The reversible phase transition is related to the atomic structure, charge, spin, and orbital dynamics (Gao et al., 2012; Zhou et al., 2013; Cui et al., 2018). The thermochromic behavior in near-infrared

wavelengths makes VO₂ an attractive candidate for smart window applications, as shown in **Figure 1A**. However, several challenges have to be addressed for large-scale applications. As the intrinsic absorption band lies in the visible region, VO₂ has a low luminous transmittance in semiconducting and metallic states. In addition, the modulation is typically limited to the near-infrared region, leading to a low ΔT_{sol} . Besides, a relatively high transition temperature of VO₂ (around 68°C) might require heating, which imposes practical limitations for use in buildings.

Many attempts have been made to enhance the thermochromic properties of VO₂. Elemental doping with high valence cations (such as W⁶⁺, Mo⁶⁺ and Nb⁵⁺) is a common way to decrease T_c by increasing the carrier concentration (Shen et al., 2021). Zhang et al. prepared W-doped VO₂ by a hydrothermal recrystallization method (Zhang et al., 2020). A 0.7 at.% W-doping film shows a suitable T_c of 42.7°C, average T_{lum} of 61.7%, and ΔT_{sol} of 11.7%. Besides, some dopants (such as Mg and Ti) can increase the T_{lum} due to the widened optical bandgap. As single doping can only improve one of the properties, co-doping with two elements has also been studied. By co-doping Hf and W into VO₂, Wang et al. modified the thermochromic properties of VO₂ alloy films with a T_c as low as 38.9°C, and a T_{lum} of 41.1% (Wang et al., 2021).

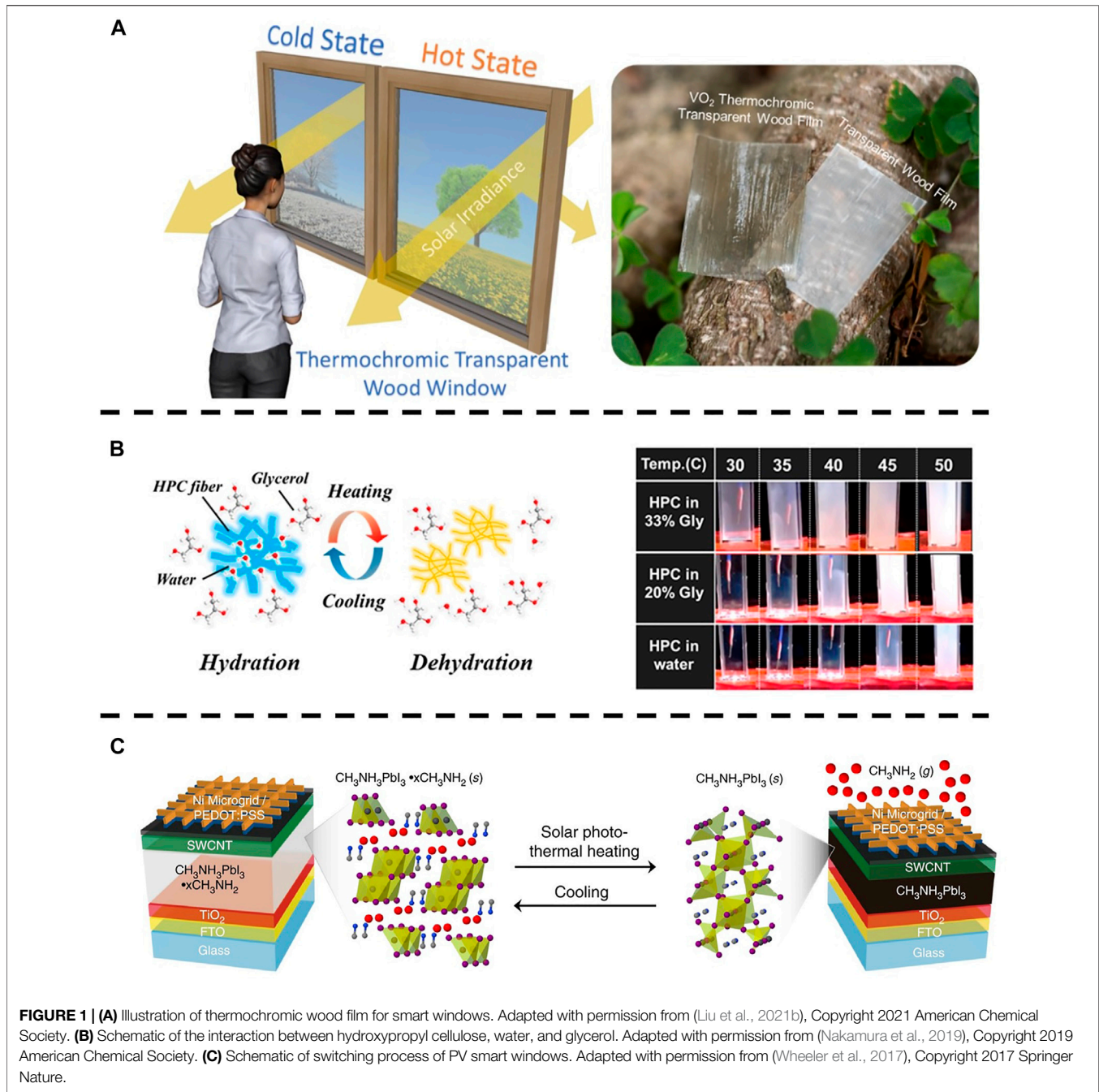
Other strategies to improve the performance of VO₂ include nano- and microscale morphology engineering (Qian et al., 2014; Kim et al., 2019; Schläfer et al., 2019; Wen et al., 2021), the use of anti-reflection coating (Zhou et al., 2016; Liu et al., 2018), porosity tuning (Kang et al., 2011), and multilayered structuring (Long et al., 2016; Sun et al., 2016; Sun et al., 2017). Fabrication complexities and costs associated with the strategies mentioned above may hinder the broader application.

Figure 1A Illustration of thermochromic wood film for smart windows. Adapted with permission from (Liu et al., 2021b), Copyright 2021 American Chemical Society. (B) Schematic of the interaction between hydroxypropyl cellulose, water, and glycerol. Adapted with permission from (Nakamura et al., 2019), Copyright 2019 American Chemical Society. (C) Schematic of switching process of PV smart windows. Adapted with permission from (Wheeler et al., 2017), Copyright 2017 Springer Nature.

HYDROGEL

Hydrogels are cross-linked hydrophilic polymer chains that swell in water but maintain defined structures. Thermochromic hydrogel undergoes a drastic transmittance change between hydrophilic and hydrophobic states around the lower critical solution temperature (LCST). At temperatures below the LCST, the intermolecular hydrogen bonds connect the polymer chains and water molecules, leading to a hydrophilic property with high transmittance. For temperatures higher than LCST, the polymer collapses due to the breakdown of hydrogen bonds and scatters light, resulting in a hydrophobic state with low transmittance.

The hydrogel is easily fabricated for smart windows with excellent solar modulation. Zhou et al. presented a thermochromic smart window using poly(N-iso-propylacrylamide) (PNIPAm) hydrogel as the active material and investigated the effect of the film thickness (Zhou et al., 2014). For a 52 μm thick film, the hydrogel shows a T_{lum}



of 87.9 and 59.9% at low and high temperatures, respectively, with ΔT_{sol} of 20.4%. The scattering behavior of PNIPAm hydrogel can be tuned by modifying the particle size, internal structure, and volume fraction of water. However, size limitations in emulsion polymerization of PNIPAm particles lead to inefficient scattering in the infrared region. To increase the hydrogel particle size and extend the transmittance modulation spectrum in the infrared region, Li et al. synthesized poly(N-isopropylacrylamide)-2-aminoethylmethacrylate hydrochloride (PNIPAm-AEMA) microparticles as a thermochromic material, which achieved a T_{lum} of 87.2% with ΔT_{sol} of 81.3% at $\sim 32^\circ\text{C}$ (Li et al., 2019). The

transmittance spectra of a 240 μm thick PNIPAm-AEMA varies dramatically when the temperature increases from 31°C to 34°C , indicating a temperature-responsive phase transition. Furthermore, device-level tests show excellent stability with less than a 4% decrease in infrared transmittance after 1,000 cycles of heating-cooling. In order to adapt thermochromism in cold weather, Nakamura et al. tuned the transition temperature of hydroxypropyl cellulose (HPC) by changing the glycerol concentration, as shown in **Figure 1B** (Nakamura et al., 2019). Note that, in this case, a rigorous encapsulation strategy is required for practical applications due to the liquid-like characteristics of the hydrogel.

TABLE 1 | Summary of various thermochromic materials.

Categories	Materials/Methods	T_{lum}^{cold} [%]	T_{lum}^{hot} [%]	ΔT_{sol} [%]	T_c [°C]	Advantages	Challenges	Refs
VO ₂	Chemical doping	61.7	—	11.7	42.7	Implementation, tunable property	Intrinsic thermochromic limitations, fabrication complexity	Zhang et al. (2020)
	Multilayer	55.4	53.9	4.3	52			Long et al. (2016)
	Antireflection coating	52.7	48.7	16.4	68			Liu et al. (2018)
Hydrogels	PNIPAm	87.9	59.9	20.4	32	Transition temperature	Encapsulation of liquid phase	Zhou et al. (2014)
	PNIPAm-AEMA	87.2	—	81.3	32	Solar modulation ability		Li et al. (2019)
Perovskites	CH ₃ NH ₃ PbI _{3-x} CH ₃ NH ₂	68	3	—	—	Integrated with PV	Encapsulation of gas phase, the safety issue of toxic gas	Wheeler et al. (2017)
	CH ₃ NH ₃ PbI ₃	85	34.3	25.5	54.4	Solar modulation ability	High transition temperature and wide hysteresis	Zhang et al. (2019)
	CH ₃ NH ₃ PbI _{3-x} Cl _x	85.2	30.3	23.7	40.1	Narrow transition hysteresis	Long-term stability	Liu et al. (2021a)
	CsPbI _{3-x} Br _x	81.7	35.4	—	105	Integrated with PV	High transition temperature	Lin et al. (2018)

Hydrogel particles with tunable scattering behavior have a promising outlook for further development. Besides, several composites can be created to form multifunctional hybrid hydrogel materials, providing better solar modulation, faster response to stimuli, and enhanced mechanical stability (Zhou et al., 2015a; Zhou et al., 2020).

PEROVSKITE

Perovskites are materials with a general chemical formula of ABX₃, where cations A and B are bonded with anion X. Perovskites have been widely applied in solar cells due to their excellent photoelectric properties (Green et al., 2014). Recently, the thermochromic properties of perovskite materials have attracted considerable attention in the smart windows field. It has been found that some halide perovskite materials, with the larger cation A usually the methylammonium ion, the smaller cation B being Pb, and the anion X being a halogen ion, exhibit a reversible phase and color change upon a natural thermal stimulus. The thermoresponsive behavior is based on the unique properties of perovskite, including crystallization, gas-induced reaction, and humidity-based hydration/dehydration.

The inverse temperature crystallization (ITC) process in hybrid perovskites was investigated by Saidaminov et al., where the solubility of perovskites in certain solvents decreases with the increase of temperature (Saidaminov et al., 2015). With a careful choice of solvent, temperature, and other parameters, single crystals of CH₃NH₃PbBr₃ and CH₃NH₃PbI₃ can be rapidly grown with controlled sizes and shapes. Following this pioneering work, De Bastiani et al. utilized the ITC process and tunable optical properties of perovskites to design thermochromic inks for smart window applications (De Bastiani et al., 2017). As the temperature rises from 25 to 120°C, the inks undergo a color change from yellow to black. However, the solvent needs to be sealed with the ink to achieve the reversible behavior, hindering wider application.

In addition, the gas-induced thermochromic behavior of perovskite has been explored. Zhou et al. reported a reversible

phase and color change of perovskite thin films from solid to liquid phase during the perovskite-gas interaction with methylamine (CH₃NH₂) gas (Zhou et al., 2015b). Wheeler et al. further demonstrated a switchable solar cell device using the low formation/dissociation energy of the methylammonium lead iodide-methylamine complex (CH₃NH₃PbI_{3-x}CH₃NH₂), as shown in **Figure 1C** (Wheeler et al., 2017). In this case, the device switches from a transparent state T_{lum} of 68% to an absorbing-colored state T_{lum} of 3%. Note that the safety issues of methylamine, including flammability and toxicity, need consideration for commercialization.

Humidity-stimulated thermochromic perovskites have also attracted interest. Halder et al. reported a hydrated lead halide hybrid perovskite CH₃NH₃PbI₃ with thermochromic behavior under the hydration and dehydration process (Halder et al., 2015). Zhang et al. developed a hydrated CH₃NH₃PbI₃ thermochromic window with a ΔT_{sol} of 25.5% and T_{lum} of 34.3 and 85% at the hot state (80°C) and cold state (25°C), respectively, after optimizing the mixing ratio and spin-coating speed during the fabrication process (Zhang et al., 2019). However, the challenges of relatively high transition temperature and large transition hysteresis need to be overcome. Liu et al. reported a hydrated CH₃NH₃PbI_{3-x}Cl_x thermochromic perovskite smart window, achieving a T_{lum} of 85.2 and 30.3% at the cold (room temperature) and hot (60°C) states, respectively, with a ΔT_{sol} of 23.7% (Liu et al., 2021a).

Further research has shown the potential of integrating both photoactivity and thermochromic features for smart photovoltaic windows applications. Lin et al. demonstrated a smart photovoltaic window using an inorganic halide perovskite, cesium lead iodide/bromide (CsPbI_{3-x}Br_x) (Lin et al., 2018). At room temperature, the transparent non-perovskite phase shows a T_{lum} of 81.7%, while at elevated temperature (150°C), the deeply colored perovskite phase exhibits low visible transparency of 35.4% with high power output. The two switchable phases of perovskite have different optical and photovoltaic efficiencies, making them promising candidates for energy savings.

CONCLUSION AND OUTLOOK

In recent years, smart windows have been intensely studied for energy-saving and indoor comfort. This mini-review focuses on the recent advancements of thermochromic materials for smart windows in terms of operation, performance, and potential for commercialization. The performances of promising thermochromic materials, including vanadium dioxide, hydrogels, and perovskites, have been summarized and compared, as listed in **Table 1**. Reference source not found. Understanding the fundamental mechanisms affecting optical properties and transition and leveraging state-of-art advancements can accelerate the development of energy-saving smart windows.

In order to fill the gap between lab-scale demonstrations and practical applications, further research is needed. This paper suggests further research along three lines: 1) Relative to conventional VO₂ materials, emerging materials like hydrogel and perovskite have shown better thermochromic performance. Further research could focus on various strategies for integration, especially for perovskites, which have shown potential for demonstrating several features beyond thermochromism; 2) Besides the three major thermochromic materials discussed in the review, exploring new materials with novel thermochromic mechanisms will help expand the range of potential candidates. For example, ionic liquids, liquid crystals (Jiang et al., 2021), and metamaterials have shown promising thermochromic properties; 3) Finally, the durability and safety of

materials should be considered, especially for materials requiring encapsulation of gas or liquid. The leakage of thermo-responsive materials may lead to a dramatic decline in performance.

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All authors listed have made substantial, direct, and intellectual contribution to the work and approved it for publication.

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