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**Title:** A Thermal Radiation Modulation Platform by Emissivity Engineering with Graded Metal-Insulator Transition

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## Abstract

Thermal radiation from a black body increases with the fourth power of absolute temperature ( $T^4$ ), an effect known as the Stefan-Boltzmann law. Typical materials radiate heat at a portion of this limit, where the portion, called integrated emissivity ( $\epsilon_{\text{int}}$ ), is insensitive to temperature ( $|\text{d}\epsilon_{\text{int}}/\text{d}T| \sim 10^{-4}/^\circ\text{C}$ ). The resultant radiance bound by the  $T^4$  law limits our ability to regulate radiative heat. Here we show an unusual material platform in which  $\epsilon_{\text{int}}$  can be engineered to decrease in an arbitrary manner near room temperature ( $|\text{d}\epsilon_{\text{int}}/\text{d}T| \sim 8 \times 10^{-3}/^\circ\text{C}$ ), enabling unprecedented manipulation of infrared radiation. As an example, we program  $\epsilon_{\text{int}}$  to vary with temperature as inverse of  $T^4$ , precisely counteracting the  $T^4$  dependence, hence thermal radiance from the surface becomes temperature-independent, allowing the fabrication of a flexible and power-free infrared camouflage with unique advantage in performance stability. The structure is based on thin films of tungsten doped vanadium dioxide where the tungsten fraction is judiciously graded across a thickness less than the skin depth of electromagnetic screening.

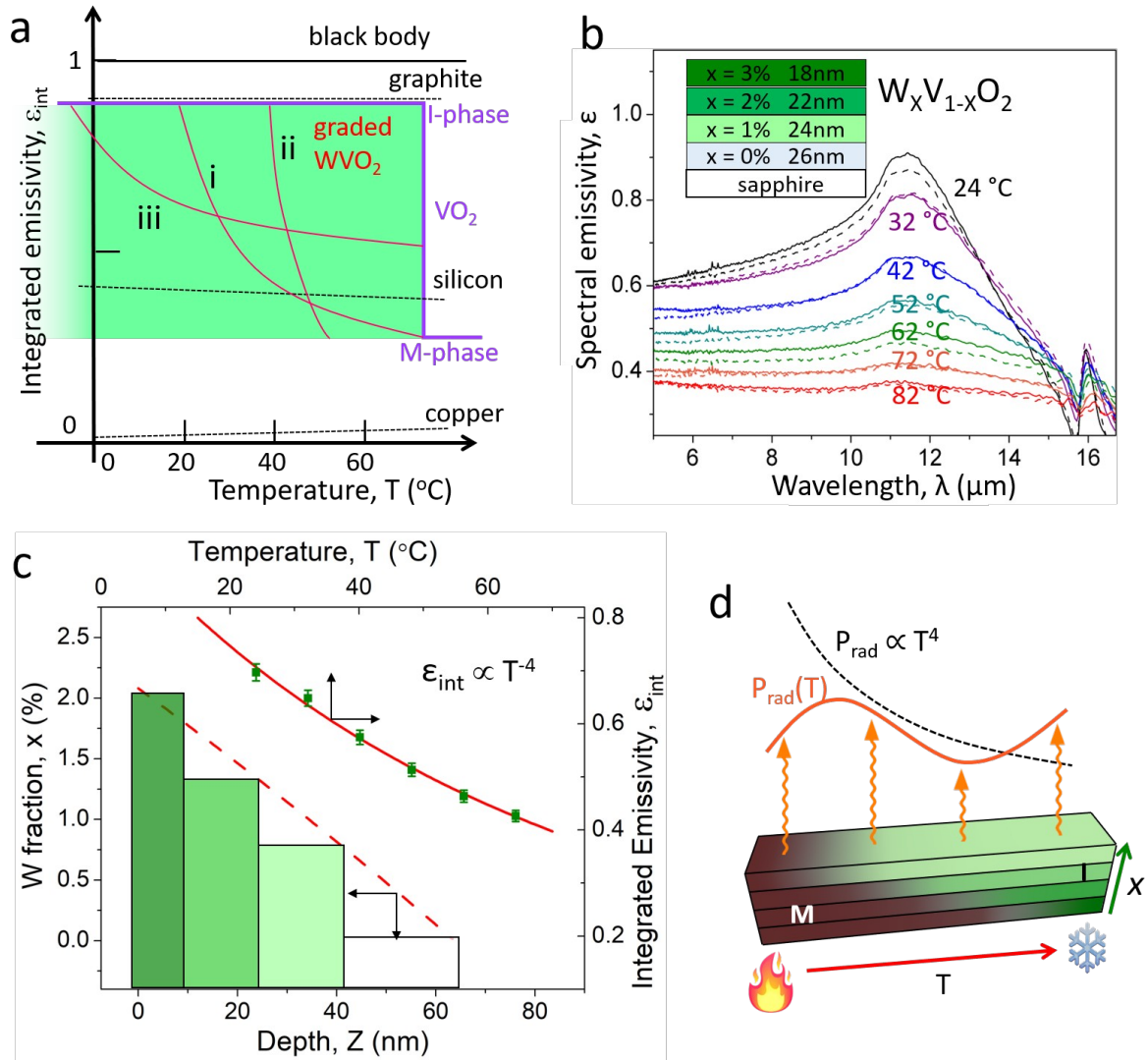
## Main text paragraphs

As a strongly correlated electron material, vanadium dioxide ( $\text{VO}_2$ ) features a well-known, temperature-driven metal-insulator phase transition (MIT) at  $T_{\text{MIT}} = 67^\circ\text{C}$  [1-3]. It takes the insulating (I) phase at  $T < T_{\text{MIT}}$  and abruptly switches to the metallic (M) phase at  $T > T_{\text{MIT}}$ . In the thermal IR spectral range near room temperature (wavelength between approximately 5 and 20  $\mu\text{m}$ ), the M phase of  $\text{VO}_2$  is much more reflective than the I phase, because its plasma energy of free electrons is  $\sim 1 \text{ eV}$  [4, 5], significantly higher than that of the thermal IR photons ( $\sim 0.1 \text{ eV}$ ). Therefore, much less IR energy is absorbed when  $\text{VO}_2$  undergoes the phase transition and becomes metallic [6-9]. According to the Kirchoff's law of radiation [10], the spectral emissivity ( $\lambda$ ) is equal to absorptivity in the IR regime. When averaged for thermal

radiation, an abrupt drop<sup>[11]</sup> in the integrated emissivity ( $\epsilon_{\text{int}}$ ) and hence thermal radiance is thus expected for VO<sub>2</sub> at  $T = T_{\text{MIT}}$ , as schematically illustrated in Figure 1a, and experimentally measured and shown in Figure S1. Here the thermal radiance per unit solid angle is given by<sup>[12]</sup>

$$P_{\text{rad}}(T) = \int_0^{\infty} B(T, \lambda)(\lambda) d\lambda,$$

where  $B(T, \lambda) = \frac{2hc^2}{\lambda^5} \frac{1}{e^{hc/(\lambda k_B T)} - 1}$  is the spectral radiance of a black body,  $k_B$  is the Boltzmann constant,  $h$  is the Planck constant,  $c$  is the speed of light,  $\lambda$  is the wavelength, and  $(\lambda)$  is the spectral emissivity. Conveniently,  $T_{\text{MIT}}$  of VO<sub>2</sub> can be rapidly shifted from 67°C to lower temperatures, down to temperatures beyond -100°C, by doping with tungsten (W) to form W<sub>x</sub>V<sub>1-x</sub>O<sub>2</sub> alloys<sup>[13-15]</sup>. The reduction of  $T_{\text{MIT}}$  follows a rate of  $dT_{\text{MIT}}(x)/dx \approx -25$  °C/at.% as measured in our films and shown in Figure S2. The contrasts in both electrical resistivity and  $\epsilon_{\text{int}}$  between the I and M phases are largely retained for low-fraction W doping. However, the abrupt nature of MIT in W<sub>x</sub>V<sub>1-x</sub>O<sub>2</sub> at those  $T_{\text{MIT}}$  values limits any application of the MIT to be within a narrow temperature window around  $T_{\text{MIT}}$ . This window is typically narrower than ~ 20°C (Figure S2), and outside this window, the material would behave as either a regular insulator (in the I phase) or a regular metal (in the M phase).



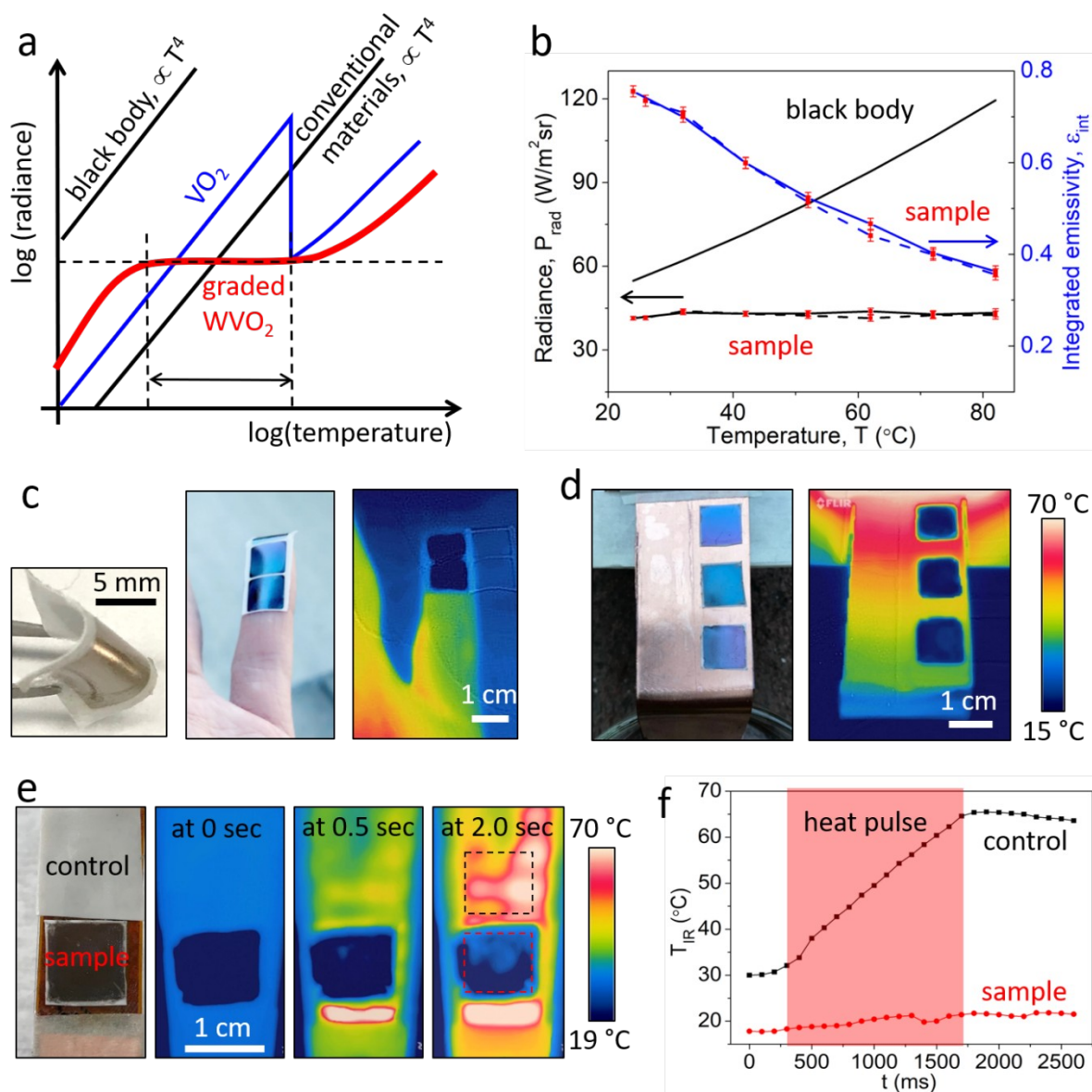
**Figure 1. Engineering thermal IR emissivity by grading metal (M) - insulator (I) transition.**

**a.** Schematic illustrating realization of nearly arbitrary temperature dependence of integrated emissivity ( $\epsilon_{int}$ ) within the shaded area using graded W-doped  $VO_2$ , represented by the three arbitrarily designed emissivity curves denoted with i, ii and iii. The behavior of typical high, moderate and low- $\epsilon_{int}$  materials and of  $VO_2$  are also shown for comparison. **b.** Spectral emissivity of a graded  $W_xV_{1-x}O_2$  structure (schematic in Inset) measured over a broad range of temperature, showing a smoothly but rapidly reduced  $\epsilon_{int}$ . Solid and dashed curves correspond to data collected during heating and cooling, respectively. **The spectral behavior in the long-wavelength region is related to the substrate (Figure S1&S11)** **c.** Design of W doping profile  $x(Z)$  (dashed line, left-bottom axes) to generate the desired  $\epsilon_{int}(T)$  relation

(solid line, right-up axes). The boxes represent actual nominal doping profile used in the  $WVO_2$  film deposition, and the data points are the experimentally measured  $\epsilon_{int}(T)$  from this film, which agrees well with the desired  $\epsilon_{int}(T)$ . **d.** Schematic showing that by rational design of the W doping profile ( $x$  along thickness direction) of  $W_xV_{1-x}O_2$ , emissivity can be programmed to regulate thermal radiation ( $P_{rad}$ ) for distinctly different behavior from the Stefan-Boltzmann  $T^4$  law.

To overcome this limit, we designed and deposited multilayer  $W_xV_{1-x}O_2$  films with total thickness less than  $\sim 100$ nm, as schematically shown in the Inset of Figure 1b. Interlayer diffusion of W took place during the film deposition and post-deposition annealing. As a result, the multilayer structure becomes a graded W-doped  $VO_2$  film, where  $x$  varies continuously across the thickness direction (Figure S2 & S3). Indeed, the lack of kinks and abrupt changes in the electrical sheet resistance of the multilayer indicates that it becomes a smoothly graded W-doped structure. The graded doping spreads out the MIT from the original narrow temperature window near  $T_{MIT}$  to a broad temperature range spanning from  $T_{MIT}(x_{max})$  to  $T_{MIT}(x_{min})$ . The drop in emissivity between the I and M phases becomes no longer abrupt and is also extended to this temperature range. We note that it is critical to grade the MIT within a thickness less than the skin depth ( $\sim 130$  nm) of electromagnetic screening in the M-phase, so that the emissivity for surface radiation can be modulated by the MIT progressing away from the film surface (details in Supplementary Information). The temperature dependence of  $\epsilon(\lambda)$  of a representative graded  $WVO_2$  film was measured using Fourier Transform IR spectroscopy (FTIR) and is shown in Figure 1b. The spectral emissivity is integrated with  $B(T, \lambda)$  over the atmospheric transparency window ( $8 \mu\text{m} - 14 \mu\text{m}$ ) and divided by the black body integration between the same wavelength limits to obtain  $\epsilon_{int}(T)$ .

The large reduction in  $\varepsilon_{\text{int}}$  from  $\sim 0.75$  to  $\sim 0.35$  over a temperature range of  $\sim 50^\circ\text{C}$  (schematic curve i in the shaded area in Figure 1a), at a rate of  $d\varepsilon_{\text{int}}/dT \sim -8 \times 10^{-3}/^\circ\text{C}$ , is orders of magnitude stronger than that of conventional materials. For example, as schematically shown in Figure 1a,  $d\varepsilon_{\text{int}}/dT$  near room temperature is about  $2 \times 10^{-5}/^\circ\text{C}$  for graphite (a high- $\varepsilon_{\text{int}}$  material)<sup>[16]</sup>,  $-1 \times 10^{-4}/^\circ\text{C}$  for silicon (a moderate- $\varepsilon_{\text{int}}$  material)<sup>[17]</sup>, and  $1 \times 10^{-4}/^\circ\text{C}$  for typical metals (low- $\varepsilon_{\text{int}}$  materials)<sup>[18]</sup>. This temperature insensitivity of  $\varepsilon_{\text{int}}$  in conventional materials arises fundamentally from the fact that temperature variation is a small perturbation in energy with respect to typical radiative electronic and phononic processes in solids<sup>[19]</sup>. The W doping profile can be designed to be more tightly distributed across the structure to achieve even greater  $|d\varepsilon_{\text{int}}/dT|$  (curve ii in Figure 1a), but at the cost of a narrower working temperature range. An example of designing the W doping profile to successfully create the desired  $\varepsilon_{\text{int}}(T)$  is shown in Figure 1c (details in Supplementary Information). Therefore, this structure provides a platform where emissivity can be engineered to exhibit nearly arbitrary, strong temperature dependence beyond that of conventional materials. Following the Stefan-Boltzmann law, the thermal radiance from the surface would be regulated to have temperature dependences distinctly different from the conventional  $T^4$  law, as depicted in Figure 1d. Exploiting the  $\varepsilon_{\text{int}}$ -regulated thermal radiation, we demonstrate a mechanically flexible and power-free thermal IR camouflage as well as a thermal IR decoy, both inherently robust and immune to drastic temporal fluctuation and spatial variation of temperature.



**Figure 2. Mechanically flexible, power-free, and  $\sqrt{T}$ - and  $dT/dt$ -immune camouflage based on the graded  $WVO_2$ .** *a* Schematic illustrating constant thermal radiance over a wide range of temperature from the camouflage, in stark contrast to the Stefan-Boltzmann  $T^4$  law from conventional materials. *b*. Thermal radiance and integrated emissivity of the film as a function of temperature, showing that the negative temperature coefficient of emissivity counteracts the  $T^4$  law to yield a temperature-independent thermal radiation power. Solid and dashed curves are for measurements during heating and cooling, respectively. *c*. Optical image of a graded  $WVO_2$  film transferred onto a PE tape showing high mechanical flexibility.



*Visible color of the sample is related to the WVO<sub>2</sub> thickness and lighting on surface (see experimental section). The fingertip is hidden from IR camera when covered with the camouflage. d. Camouflaging an object with large spatial temperature variation from ~25°C to 65°C, showing immunity to  $\nabla T$ . e. Camouflaging an object with drastic temperature surge, showing immunity to  $dT/dt$ . As shown in f, the actual temperature of the camouflage (represented by the IR temperature of the control which is a high-emissivity tape placed near the sample) rises rapidly by the heating pulse, while the IR temperature of the camouflage stays nearly flat. The IR temperature was measured by averaging the camera reading in the center  $1 \times 1$  cm<sup>2</sup> region of each film (dashed boxes), and the error bars are comparable to the symbols in size.*

As the first example, we demonstrate a thermal IR camouflage using the graded WVO<sub>2</sub> platform, of which the working principle is shown in Figure 2a. When  $x$  and thickness of each layer were judiciously selected (Figure 1c), the graded doped WVO<sub>2</sub> exhibits an  $\epsilon_{\text{int}}$  that depends on temperature following  $\sim 1/T^4$ . The FTIR-measured  $\epsilon_{\text{int}}(T)$  of a sample prepared following this design method (details in Figure S4) is shown in Figure 2b. The  $1/T^4$  dependence of  $\epsilon_{\text{int}}(T)$  is multiplied with the Stefan-Boltzmann  $T^4$  relation of a black body, to give the  $T$ -independent thermal radiance  $P_{\text{rad}}$ , as shown in Figure 2b. The constant  $P_{\text{rad}}$  over a temperature range over 50°C near room temperature invalidates the assumption of  $T$ -dependent  $P_{\text{rad}}$ , the principle behind IR imaging<sup>[20, 21]</sup>. As such, the structure would appear to have a constant IR temperature ( $T_{\text{IR}}$ ) when imaged by an IR camera, despite the reality that its actual temperature ( $T_{\text{actual}}$ ) may vary widely. Unlike low-emissivity camouflage coatings that rely on reflection signals from a confinement and thus limited to indoor scenarios, the IR

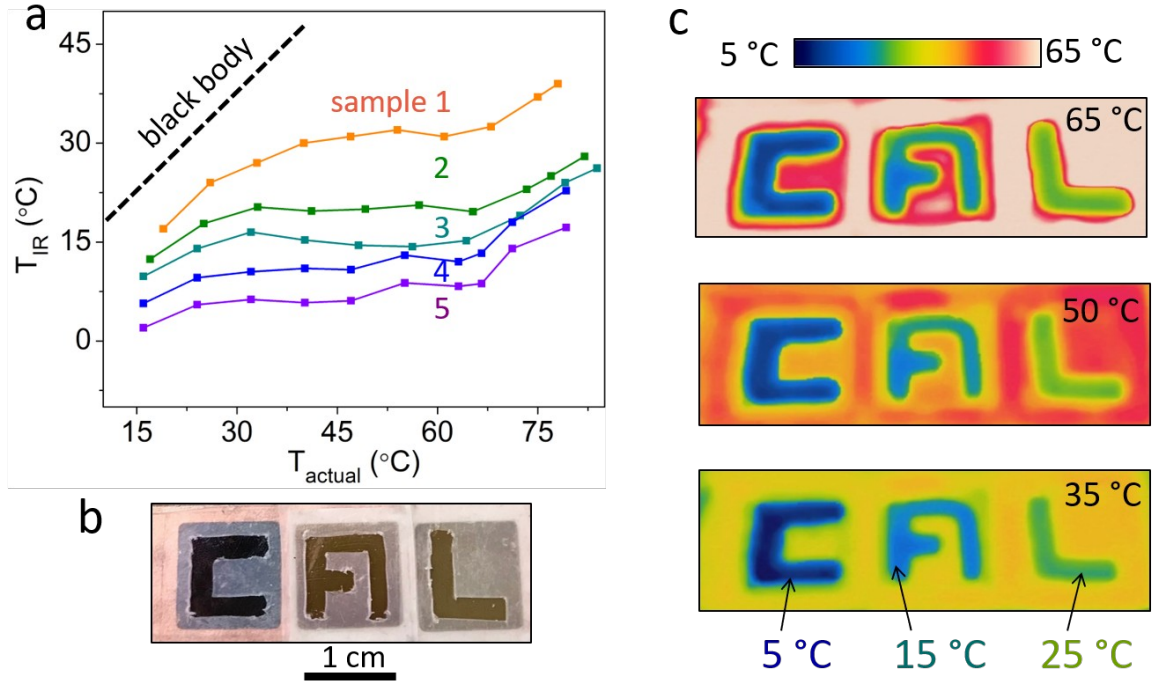
camouflage based on graded WVO<sub>2</sub> fools the camera by manipulating thermal radiation from the target itself, making it ideal for outdoor applications.

Existing IR camouflage techniques use a coating whose emissivity can be modulated by effects of thermochromics<sup>[22-24]</sup>, electrochromics<sup>[25-28]</sup>, photocarrier doping<sup>[29]</sup>, or mechanical actuation<sup>[30]</sup>. These active camouflage techniques generally require energy input to operate, and are based on rigid and complicated heterostructures. They are also typically limited to only camouflage objects with uniform and slow-varying temperature profiles. In contrast, our graded-WVO<sub>2</sub> offers an IR camouflage that is power-free, monolithic, and mechanically flexible. A detailed comparison of key capabilities and figures-of-merits with existing IR camouflage systems can be found in Table S3. Using a wet etch-transfer method (see Methods and Figure S5), the graded-WVO<sub>2</sub> film is transferred from the growth substrate onto a polyethylene (PE) tape. As the total thickness of the WVO<sub>2</sub> film is less than 100 nm, the WVO<sub>2</sub>/tape structure can be bent to curvatures exceeding 10/cm (Figure 2c) without degrading its camouflage performance. The mechanical flexibility combined with the angular-independence of emissivity (Figure S6) allows the camouflage to be easily applied as a tape onto non-flat surfaces, with minimal or no impact to the performance (Figure S7). No noticeable degradation in camouflage property or change in surface morphology was detected after up to 100 heating/cooling cycles through the working temperature range (15-65 °C), demonstrating good reliability of the film (Figure S8). In Figure S8, A  $T_{IR}$  difference of ~ 3 °C is observed between the heating and cooling branches, caused by the hysteresis effect of the WVO<sub>2</sub> phase transition. This degree of impact should not affect the general applicability of the technique, and could be reduced by future work to optimize the hysteresis, including process by crystal dehydration<sup>[31]</sup> or Ti doping method<sup>[32]</sup>.

A series of experiments were conducted to test the performance of the camouflage. Figure 2c shows optical image and IR snapshot of a finger where the tip is covered by the camouflage. Distinct from the exposed skin, the part covered by the camouflage displays a  $T_{\text{IR}}$  similar to the environment, concealing the fingertip from IR detection. Apart from such conventional application as a living body camouflage, our structure has the inherent, unique advantage to cover up objects with large spatial variation ( $\nabla T$ ) or abrupt temporal fluctuation ( $dT/dt$ ) of temperature. In Figure 2d, a copper plate coated with high-emissivity tape is thermally biased with  $T_{\text{actual}}$  varying from 25 °C to 65 °C. Three identical WVO<sub>2</sub> camouflages were placed at different positions of the plate that have distinct local temperatures. The regions underneath the camouflages are simultaneously masked from IR detection regardless of the different local temperatures and high thermal gradient. Compared to using multiplexing to address the spatial temperature variation in conventional camouflages<sup>[26, 29]</sup>, the graded WVO<sub>2</sub> design offers a much simpler, monolithic, power-free and tetherless approach.

The camouflage is also inherently immune against abrupt temperature fluctuation. In Figure 2e, a heater membrane is partly covered by our graded WVO<sub>2</sub> film transferred onto a high-emissivity PE tape (sample), and the rest is covered by the high-emissivity PE tape alone (control). The temperature of the plate shoots up rapidly when a current pulse passes through the heater. The IR camera catches clearly a temperature spiking up to 65 °C within less than 2 seconds from the control surface, while the camouflaged surface stays at nearly constant  $T_{\text{IR}}$  of 20 °C irrespective of the heat pulse (see video in Supplementary Information, Video S1). The detected  $T_{\text{IR}}$  is plotted as a function of time in Figure 2f, which shows a nearly total immunity to the temperature surge. This immunity is a substantial advantage over the strategy of adjusting the emissivity using a feedback loop in conventional camouflages, as the latter typically has a response time over seconds<sup>[26, 27, 30]</sup> and would have the object exposed to detection during the response lagging time.

Next, we demonstrate another unique function, a thermal IR “decoy”, using the structure. A decoy is different from the camouflage in the way that the decoy would not only passively conceal from IR camera the real thermal activity of the object, but also intentionally fool the camera with a counterfeited  $T_{\text{IR}}$  image that is independent of  $T_{\text{actual}}$  distribution and fluctuation. That is, to create a robust thermal radiative pattern  $T_{\text{IR}}(X, Y, Z)$  on a surface that has arbitrary actual temperature distribution  $T_{\text{actual}}(X, Y, Z, t)$ , where  $X, Y$  and  $Z$  are the coordinates of the surface and  $t$  is time. Because the overall emissivity of the graded-WVO<sub>2</sub>/tape system is the combined effect of both the WVO<sub>2</sub> film and the tape, by using different doping profiles for the graded WVO<sub>2</sub> film and transferring onto different tapes,  $T_{\text{IR}}$  of the camouflage can be designed to take distinct values to meet different requirements. A series of flexible WVO<sub>2</sub> / tape samples were prepared (details in Figure S9). As shown in Figure 3a, despite  $T_{\text{actual}}$  varying widely from room temperature to  $\sim 70^\circ\text{C}$ , these samples all exhibit a nearly constant  $T_{\text{IR}}$  within the  $T_{\text{actual}}$  range, whereas  $T_{\text{IR}}$  is designed to be equal to any desired value in the range of  $5^\circ\text{C} \sim 30^\circ\text{C}$ . As a proof of concept, Figure 3b and Figure 3c demonstrate a working decoy where the image of “CAL” shows a stable IR temperature reading, regardless of the change in  $T_{\text{actual}}$ . Such decoying information can be also encoded during the film deposition by laterally patterning the composition and thickness of the graded WVO<sub>2</sub> film, allowing fabrication of a monolithic and lithographically manufacturable decoy (Figure S10).



**Figure 3. Demonstration of  $T_{actual}$ -independent thermal IR decoy.** *a.* IR temperature ( $T_{IR}$ ) versus actual temperature ( $T_{actual}$ ) of five flexible structures, where the graded  $WVO_2$  configuration is designed to yield  $T_{actual}$ -independent, but different values of, desired  $T_{IR}$  within the working range of  $T_{actual}$ . Details of these samples are shown in Fig.S9. The error bars are comparable to the symbols in size. *b* (optical) and *c* (IR images), using the sample 1, 3 and 5, respectively, a pattern of “CAL” is made to show “C” at constant  $T_{IR} \approx 5^\circ\text{C}$ , “A” at  $T_{IR} \approx 15^\circ\text{C}$  and “L” at  $T_{IR} \approx 25^\circ\text{C}$ , regardless of  $T_{actual}$  varying to 65, 50 or 35 °C. The three samples are all  $1 \times 1 \text{ cm}^2$  in size.

While these features greatly expand the application of radiative camouflage to inhomogeneous and dynamic environments as well as IR decoying, they also inspire novel device concepts based on reflection or transmission of IR signals. As expected, over the broad spectral range from near-IR (down to the plasma wavelength  $\sim 1.2 \mu\text{m}$ ) to the far-IR, the reflectivity of the graded  $WVO_2$  would exhibit a temperature dependence that is much

stronger than that of conventional materials (Figure S11 shows part of the spectrum). This feature enables potential ways for high-sensitivity remote temperature sensing using thermorefectance imaging, or active reflectance modulation of IR signals. Therefore, the graded  $\text{WVO}_2$  structure offers a general platform for unprecedented manipulation and processing of IR signals for novel radiative and reflective systems.

## Experimental Section

**Thin film deposition and etching/transfer process.**  $\text{WVO}_2$  thin films were grown on c-cut sapphire and quartz substrates using pulsed laser deposition (PLD). The graded W doping was achieved by sequentially focusing the pulsed laser on a series of  $\text{WVO}_2$  targets with different W doping ratios. The targets were prepared by mixing  $\text{WO}_3$  and  $\text{V}_2\text{O}_5$  powders with W:V atomic ratio ranging from 0 to 3.0%, then made into 1 inch diameter round discs with a hydraulic press. All thin films were deposited in 5 mTorr  $\text{O}_2$  environment at 475 °C substrate temperature. The PLD laser energy was set at 321 mJ with 5 Hz pulse frequency. A post-deposition anneal at 475 °C for 30 mins in the same 5mTorr  $\text{O}_2$  environment was performed for all graded doped  $\text{WVO}_2$  film. The thickness and deposition rate of the thin films were measured by atomic force microscopy and scanning electron microscopy (Figure S12), and the W fraction as well as depth profile were characterized by x-ray photoelectron spectroscopy (Figure S3).

$\text{WVO}_2$  thin films grown on quartz substrates were transferred onto PE tapes by sticking the  $\text{WVO}_2$  side to the adhesive side of the tape, and then etching off the 170  $\mu\text{m}$  thick quartz substrate by dipping into 49% Hydrofluoric (HF) acid for 5 mins. The samples with transferred  $\text{WVO}_2$  were then rinsed in deionized water for 3 mins and gently blown dry with a  $\text{N}_2$  gun. The interfacial adhesion is strong and no delamination of  $\text{WVO}_2$  is observed during the wet transfer process, in further contact with water, or with mechanical bending and twisting of the tape. The thickness of the PE tape (default) and the scotch tape is ~ 0.20 mm and 0.08 mm, respectively. The visible color of mainly depends on the thickness of the graded  $\text{WVO}_2$  coating layer. Thicker film will appear blue/purple, while thinner film tends to have brown/yellow color. When taking a photo, the lighting and the reflection will affect the visible appearance of the sample surface as well.

## FTIR measurements and IR imaging

The normal-direction emissivity  $\varepsilon(\lambda, T)$  was obtained from normal-incident reflection measurements following the equation:  $\varepsilon(\lambda, T) = 1 - R(\lambda, T)$ , due to negligible transmission  $T(\lambda, T)$  of the sample over the spectra of interest. The reflection spectra,  $R(\lambda, T)$ , were characterized by an Agilent Cary 670 FTIR spectrometer and Agilent Cary 620 microscope system with a 15× objective lens (numerical aperture of 0.62). A blade aperture of  $60 \mu\text{m} \times 60 \mu\text{m}$  was used to select the area of interest. For measurements of samples on PE tapes, flat regions away from substrate curving spots were chosen to minimize scattering, and multiple ( $> 3$ ) measurements were performed to confirm repeatability of the results. All reflection spectra were normalized to the reflection spectrum of a 300 nm thick gold film. The temperature of the samples was controlled by a customized closed-loop thermal stage, connected to a Lakeshore 321 temperature controller.

The IR images and videos were captured by a FLIR ONE infrared camera. To avoid reflection signals from the camera and the surrounding, the default viewing angle was set as  $15^\circ$  instead of normal incident direction, and the experiments were performed in an **open-area outdoor environment under clear sky (little cloud)**. The temperature was controlled by the same thermal stage used in the FTIR measurements. When taking an IR image, the camera measures the incident thermal radiation  $P_{\text{rad}}$ , and gives the temperature reading ( $T_{\text{IR}}$ ) assuming a constant emissivity for the target ( $\varepsilon_0 = 0.90$ ). The relationship can be described by the equation:  $\varepsilon_0 \sigma T_{\text{IR}}^4 = P_{\text{rad}}$ . The  $T_{\text{IR}}$  was then plotted as a function of actual temperature ( $T_{\text{actual}}$ ) to demonstrate the camouflage performance.

**The error bars for data presented in this work are based on standard deviation of at least 3 separate samples. This applies to both the figures in the manuscript and the supporting information.**



### **Conflict of interest**

The authors declare no conflict of interest

### **Supporting Information**

Supporting Information is available from the Wiley Online Library or from the author.

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### **Author Contributions**

J.W. conceived the project. J.W. and K.T. designed the experiments. K.T. prepared the materials, fabricated the devices and performed the measurements. X. W. helped with training and measurements of the FTIR. X. Z. provided instrumental support for the FTIR

experiments. K. D. performed theoretical modeling. J. L. helped with preparation of the figures and SEM measurement. All authors contributed to discussing the data and editing the manuscript.

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