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Femtosecond laser based “Janus-material/function” processing paradigm for photothermal and radiative cooling modulation

Jianing Liao¹, Zhuguo Li^{1,2*} and Dongshi Zhang^{1*}

Abstract: The “Janus” concept, integrating asymmetric functions like radiative cooling (RC) and photothermal (PT) effects is gaining prominence for multi-scenario applications and function enhancement. In this work, we present a novel “Janus-material/function” processing paradigm by femtosecond laser nanomaterial synthesis and 600 °C thermal post annealing. This technique enables universal preparation of PT-active surfaces and in situ colorful RC functional transitions on transition metals (Nb, Mo, Ta, and W), while Ti-based PT interfaces exhibit uniquely thermal resistance. Using custom-built systems, the PT/RC effects of as-prepared “Janus” black/white PT/RC-WO_{3-x} interfaces are verified, with interfacial temperatures 2 °C lower and 9 °C higher, respectively, than that of the benchmark tungsten plate under 1-sun irradiation. These interfaces demonstrate good thermal/mechanical stabilities, withstanding 4 h of 100 °C/200 °C annealing, vigorous hand shaking, and tape peeling. Femtosecond laser in situ PT-WO_{3-x} patterning on high-thermal-conductivity, high-infrared-reflective W-substrate demonstrates high-contrast infrared display in the emission and reflection modes by 80 °C sample heating and hot water excitation, respectively; whereas annealed white/dark-blue RC-PT-WO_{3-x} matrix, characterized by low thermal conductivities, exhibits convergent low-contrast infrared display. The flexibility of this paradigm, enabling convenient integration of double-sided “Janus” PT-RC functionalization and single-sided PT-RC “LASER JANUS” patterning on a single refractory metal, highlighting its potential for advanced Janus-conceptual customization in different thermal regulation scenarios.

Keywords: Janus matrix; femtosecond laser synthesis; photothermal; radiative cooling; photo-thermoelectric generation

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1 Introduction

The Janus architecture, deriving from the dual-faced Roman deity Janus, is initially proposed in science by P. G. de Gennes in his 1991 Nobel Lecture¹. Since then, the Janus concept has been widely applied to the design of functional particles², interfaces³, and devices⁴, embodying a paradigm of asymmetric structural and physicochemical properties. Janus interfaces have ushered in a new era of advanced thermal management for applications, as exemplified by thermoelectric generation (TEG)⁵, and electronic devices heat dissipation⁶ by integrating low/high thermal conductivity properties⁷ or combining photothermal/radiative-cooling

(PT/RC) functions^{8,9}. Tungsten (W) and its sub-stoichiometric oxide (WO_{3-x}) represent a promising, yet underexplored material system for such “Janus” functionalization. The stark contrast in their thermal conductivities, ~170 W/mK for W versus 1–2 W/mK for WO_{3-x}¹⁰, enables efficient dual-mode thermal management. This stark contrast is also conducive to realize sharp-contrast infrared (IR) display¹¹ and IR camouflage¹². Furthermore, tungsten exhibits high reflectivity in the mid-infrared (MIR, 2.5–25 μm) band^{13,14}, while its Janus thermal counterpart, high-MIR-emissive WO_{3-x}, allows for direct heat dissipation into the outer space through the atmospheric transparency window (8–13 μm, in the long wavelength infrared (LWIR) band).

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Solar reflectance/absorbance in the 0.25–2.5 μm band (covering ultraviolet (UV), visible (Vis) and near-infrared (NIR)) offers another “Janus” thermal regulation route: enhancing the RC or triggering the PT effect. Reflective white surfaces with high thermal emissivity facilitate passive radiative cooling (PRC) for extreme-condition usages. PRC¹⁵ is considered to be a critical sustainable pathway to reduce cooling energy consumption¹⁶, mitigate CO₂ emissions, and support carbon-neutral initiatives¹⁷, which in turn drive the technological innovation in RC-correlated fields¹⁸. In contrast, black and colorful solar absorbers with high LWIR emissivity inherently exhibit Janus-type PT/RC duality¹⁹. Although mutual interference between opposite functions may constrain the individual performance, their integration enables continuous, temperature-adaptive thermal management²⁰ and facilitates year-round energy conservation²¹. Achieving such a PT/RC-material/functionality Janus matrix demands innovative processing paradigms that transform hard-to-process refractory tungsten into phase-tunable, multicolored WO_{3-x} with spatial patterning capacities.

Femtosecond (fs) laser processing has established a robust paradigm for producing black metal surfaces as exemplified by stainless-steel²², zirconium²³ and aluminum²⁴ and copper²⁵, with potential applications in IR camouflage²⁶. Owing to tungsten’s high melting temperature of 3422 °C²⁷, fs laser engineering typically results in laser induced periodic surface structures (LIPSS) and nanoparticles (NPs)¹⁴. LIPSS are classified into high-spatial-frequency LIPSS (HSFL) and low-spatial-frequency LIPSS (LSFL)²⁸, which can be transformed by tailoring processing parameters. To overcome hard-processing limitations, an integrated subtractive/additive-integrated fs laser manufacturing technique has been developed¹³. This method enables synthesis, in situ deposition, and flexible patterning of black WO_{3-x} nanomaterials, yielding PT-black absorbers with broadly modifiable IR emissivities. This advancement opens new opportunities for visible information encryption and differential IR decryption display²⁹. In contrast, fs laser based “white-engineering” remains relatively underdeveloped, despite the successful synthesis of white ZrO₂, grey-white Al₂O₃ and blue-white TiO₂ aerogels via fs laser processing of their respective metals, and the production of white WO_{3-x} from yellow WO₃ crystals³⁰. Although fs laser oxidative engineering readily generates colorful TiO_{2-x} and Ti-TiO_{2-x} Janus structures³¹, titanium’s relatively low thermal conductivity (21.9 W/(m·K)) and melting temperature of 1668 °C reduce its appeal compared to tungsten-based systems. Notably, electron spin resonance (ESR) studies indicate that white WO_{3-x} possess a higher density of oxygen vacancies than yellow WO₃. This suggest that oxygen vacancy engineering could offer a viable pathway for the synthesis and patterning of white WO_{3-x} structures using alternative WO_{3-x} precursors.

In this work, a novel metal-based Janus-system processing paradigm is presented, initiating with the synthesis and

in situ deposition of black, high-LWIR-emissivity WO_{3-x} nanomaterials (Fig. 1(a)), followed by a transition to a white phase upon annealing at 600 °C. The selection of 600 °C as the optimal PT-to-RC transition temperature was determined through systematic screening of samples annealed at 300 °C, 400 °C, 500 °C, 600 °C, 700 °C and 800 °C for 2 h, as shown in the optical images at the bottom of Fig. 1(a). Specifically, sample (5 W-50 mm/s) processed with a laser power of 5 W and a scan speed of 50 mm/s retains its original appearance at 300 °C, turns light blue at 400 °C, lightens significantly at 500 °C, becomes white at 600 °C, and evolve into yellowish at 700 °C and 800 °C. Time-dependent annealing at 400 °C (ranging from 5 min, 15 min, 30 min, 1 h, and 2 h) reveals a progressive dark-to-light-blue color transition. Generally, annealing at higher temperatures facilitates sufficient 3D interconnection of NPs. To optimize the structure, we comparatively analyzes pristine W, fs laser prepared LIPSS, LIPSS/NPs, and hairy hierarchical structures annealed at 600 °C and 800 °C. The phase evolution, oxygen vacancy density, ultrabroadband reflectance (absorbance) of black WO_{3-x} NPs and white WO_{3-x} structures are characterized, using the 600 °C-annealed tungsten as a reference. The passive PT and RC effects of black and white WO_{3-x} using custom-built setups, along with Janus patterning and transformation, are both demonstrated (Fig. 1(b, c)). Based on these results, potential Janus structures for various TEG scenarios are proposed, and the manufacturing capacity of double-sided and single-sided integrative PT/RC-black-white interfaces on a single tungsten plate is validated. We also demonstrate Janus matrix patterning and IR display under different external stimuli, analyzing the underlying mechanism. Finally, the universality of this fs laser based “Janus-material/function” processing paradigm was investigated by extending it to other refractory metals, including Ta, Mo, Nb, and Ti, thereby elucidating the material dependence. The advantages and limitations of this method and the resulting interfaces are briefly discussed.

2 Results and discussion

2.1 PT-to-RC transition and PT/RC materials analysis

Figure 1(d) illustrates the morphological characteristics of black WO_{3-x} NPs synthesized by fs laser ablation of W in air, as well as the white and yellow WO_{3-x} nanostructures derived from 600 °C and 800 °C annealing, respectively. The as-synthesized black WO_{3-x} NPs exhibit a broad size distribution, primarily between 20 and 40 nm. After 600 °C annealing, the NPs become interconnected with only a slight increase in structural dimension (~40 nm), indicating that the process preserves morphological integrity through delicate synergistic sintering rather than extensive particle coalescence. This stands in sharp contrast to the work of Huelser et al.³², who synthesized white WO₃ using a

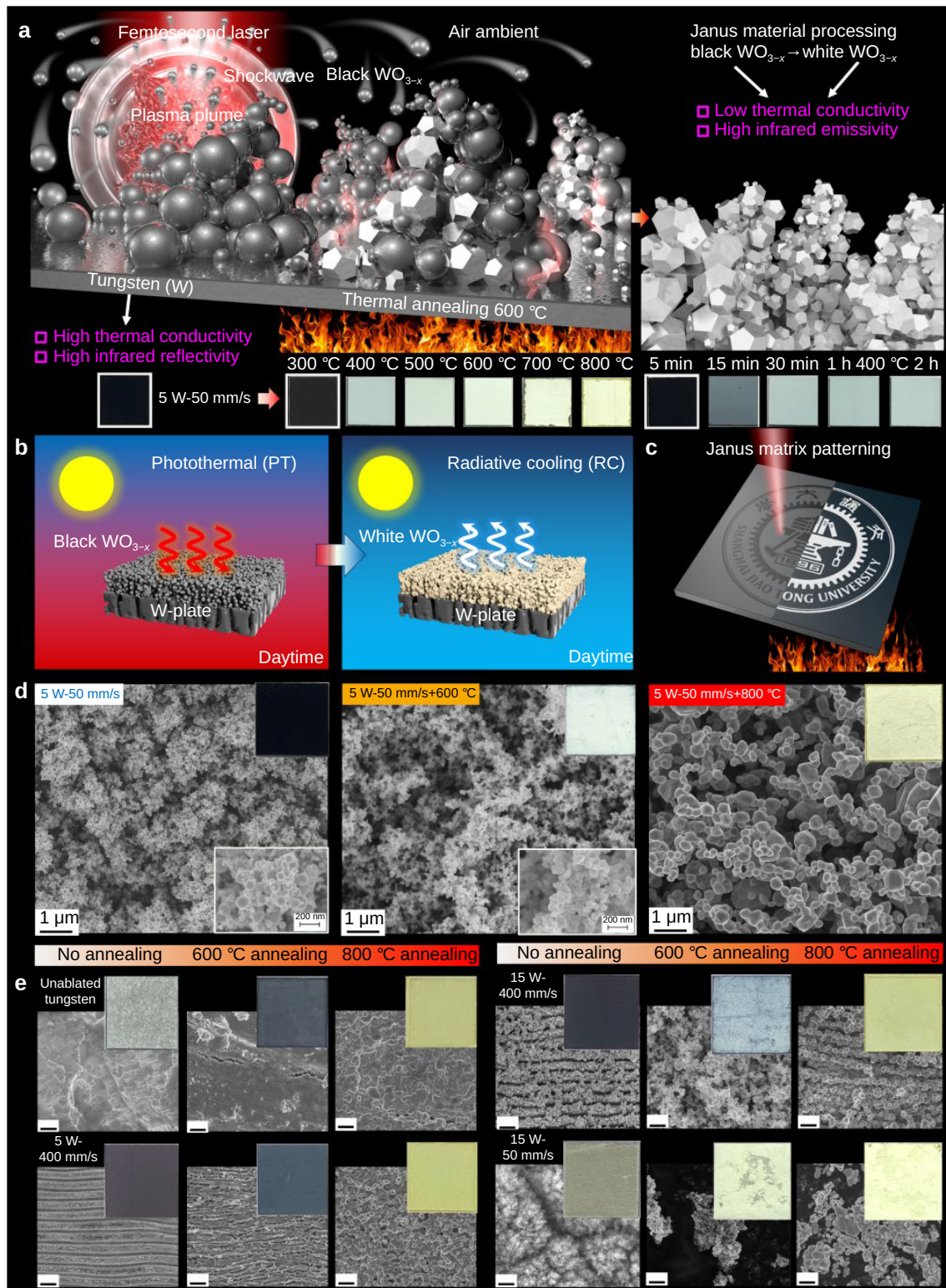


Fig. 1 | Fs laser-based Janus processing and characterization (a) Schematic of synthesis of black PT- WO_{3-x} NPs by fs laser ablation of a pure tungsten plate (high thermal conductivity, high IR reflectivity) and converting into white RC- WO_{3-x} hierarchical structures via 2h annealing at 600 °C. Optical images of as-prepared PT sample (5 W-50 mm/s) and counterparts obtained by 2h-annealing at temperatures from 300 °C to 800 °C. Color evolution upon 400 °C annealing for 5 min, 15 min, 30 min, 1 h, and 2 h. PT samples are white-framed for contrast. (b) Schematic of PT-to-RC Janus material/function transition. (c) Janus matrix patterning of the badge of Shanghai Jiao Tong University. (d) SEM and optical images of black PT- WO_{3-x} NPs, corresponding nanostructures annealed at 600 °C and 800 °C. (e) Morphology comparison of pure tungsten, LIPSS (5 W-400 mm/s), LIPSS/NPs (15 W-400 mm/s), hairy hierarchical structures (15 W-50 mm/s) and those obtained after 2h-annealing at 600/800 °C. Scale bar: 1 μm .

complex low-pressure $\text{H}_2/\text{O}_2/\text{Ar}$ flat flame with different concentrations of tungsten hexafluoride WF_6 . Our paradigm establishes a streamlined, convenient pathway for Janus-material/function processing. By utilizing inexpensive bulk metal targets, our method circumvents the complexity, high-cost chemicals, and overcoming the inherent limitation of lacking direct spatial patterning capability of chemical methods^{33,34}. Upon 800 °C annealing, the material transforms into yellow crystals with an average grain size of ~200 nm, reflecting significant grain growth and a phase transition toward stoichiometric WO_3 ³⁵.

For comparative analysis, other precursors including pristine tungsten, LIPSS (5 W-400 mm/s), LIPSS/NPs (15 W-400 mm/s), and hairy hierarchical structures (15 W-50 mm/s) were prepared by fs laser ablation and subsequently annealed at 600 °C and 800 °C. The results (Fig. 1(e)) demonstrate that densely deposited particles are the most effectively precursor for forming white WO_{3-x} . Upon 600 °C and 800 °C annealing, pure tungsten and LIPSS develop blue and yellow oxide films or grains, respectively. In contrast, low-density NPs on LIPSS induce the formation of very thin white WO_{3-x} layer; however, this layer is insufficient to mask the blue color of the underlying non-stoichiometric oxidized LIPSS. The hairy structures yield a thicker light-green film with segregated grains after 600 °C annealing, which transitions to a lighter yellow film at 800 °C. These findings further support the conclusion that NPs are superior precursor for achieving white and light-yellow WO_{3-x} phases. The LIPSS structures are partially and completely erased upon annealing at 600 °C and 800 °C, indicating the oxidative grain growth behavior and the high thermal-oxidization susceptibility of tungsten.

Figure 2(a, b) presents transmission electron microscopy (TEM), selected area electron diffraction (SAED), and high-resolution transmission electron microscopy (HRTEM) images of fs laser synthesized black NPs and the white nanostructures obtained after 600 °C annealing (exfoliated from the surface). Figure 2(c, d) displays the corresponding X-ray diffraction (XRD) and X-ray photoelectron spectroscopy (XPS) results. The black NPs are confirmed to comprise multiple phases, including monoclinic WO_3 (Pc), W_3O , metallic W, and monoclinic WO_3 (P_{21}/n). The XPS W4f spectra further confirm the incomplete oxidation of the black NPs, exhibiting distinct peaks of W^0 , W^{5+} , and W^{6+} states (Fig. 2(d)). Notably, the formation of metastable WO_3 (Pc) phase is attributed to the extreme high-pressure, high-temperature conditions and the ultrafast quenching rate (10^{12-14} K/s) inherent to fs laser ablation³⁰. Following thermal annealing at 600 °C, the structural phases transform primarily into WO_3 (P_{21}/n) (PDF#83-0950) and $\text{WO}_{2.625}$ (PDF#81-1172), which account for 59.9% and 38% of the material, with a residual 2.1% metallic W (Fig. 2(e)). The proportion of WO_3 (P_{21}/n) in the white nanostructures is significantly higher than that obtained in 600 °C-annealed pristine-W (25.9%) or LIPSS (21.7%). This enhancement is

owing to the larger surface area of the precursor NPs (Fig. 1(d)), which facilitates a higher oxidation rate. XPS spectra indicate that while metallic W is absent in both 600 °C-annealed blue (pristine W derived) and white WO_{3-x} (black NPs derived) samples, oxygen vacancies remain present. Electron paramagnetic resonance (EPR) characterization (Fig. 2(f)) reveals that the density of oxygen vacancies in white WO_{3-x} is higher than black WO_{3-x} , as indicated by the signal peaks around $g = 2.003$ ³⁶. This finding contrasts with a previous report³³ showing that white WO_{3-x} prepared by oxidative calcination and reductive calcination of WCl_6/PVP nanofiber in O_2/air possesses a lower oxygen vacancy density than its black WO_{3-x} counterpart prepared in the H_2/Ar atmosphere. This discrepancy arises because our black WO_{3-x} synthesized by fs laser ablation of W in air, retains a significant amount of metallic W and low-oxidation W_3O phases (Fig. 2(c)), rather than being sufficiently oxidized.

The coloration of WO_{3-x} is generally attributed to the lattice distortion of WO_3 induced by electrons trapped in oxygen vacancies³⁷. Alternatively, the polaron model suggests that excess electrons (introduced by doping, photo-generation, or oxygen vacancy engineering) localize at W^{6+} sites to form W^{5+} polarons³⁸. Although the precise origin of color centers for WO_{3-x} remains debated, the correlation between blue/brown colors and specific ranges of the oxygen vacancy concentration is commonly asserted³⁹. Annealing at 800 °C leads to complete oxidation of all samples, resulting in a 100% phase transition to WO_3 (P_{21}/n) (Fig. 2(c)). Figure 2(g) schematically illustrates this phase transition from black NPs to white WO_{3-x} , probably including the metastable-to-stable phase transition from WO_3 (Pc) to WO_3 (P_{21}/n)³⁰, the conversion of W_3O to $\text{WO}_{2.625}$ ^{40,41}, and the transformation of mixed W/ WO_3 phases into $\text{WO}_{2.625}$ ⁴² followed by further oxidation to WO_3 (P_{21}/n). Based on the evolution of LIPSS and pure W oxidation (Fig. 2(e)), $\text{WO}_{2.625}$ is deemed to be an intermediate phase in the oxidation pathway from W to WO_3 .

2.2 Optical, thermal regulation, and photothermoelectric evaluations

Figure 3(a–b) displays the UV-NIR and MIR spectra of black WO_{3-x} NPs, white/yellow WO_{3-x} structures, pure W, and W-annealed WO_{3-x} interfaces. The absorbance in Vis, solar spectrum, LWIR and MIR bands is quantified in Fig. 3(c). The black NPs exhibits a Vis-absorbance of 95.7% and a solar spectrum absorbance of 98%, with an LWIR emissivity of 87.6%. After white structuring, the Vis and solar spectrum absorbance decreases to 32.7% and 69.2%, respectively, while the LWIR emissivity increases to 91.5%. Based on calculations mapped onto the CIE 1931 color chromaticity diagram (Fig. 3(d)), the color of the white WO_{3-x} closely resembles that of WO_{3-x} aerogels synthesized via fs laser-bionic manufacturing of yellow WO_3 grains³⁰. This similarity addresses the interface integration challenge

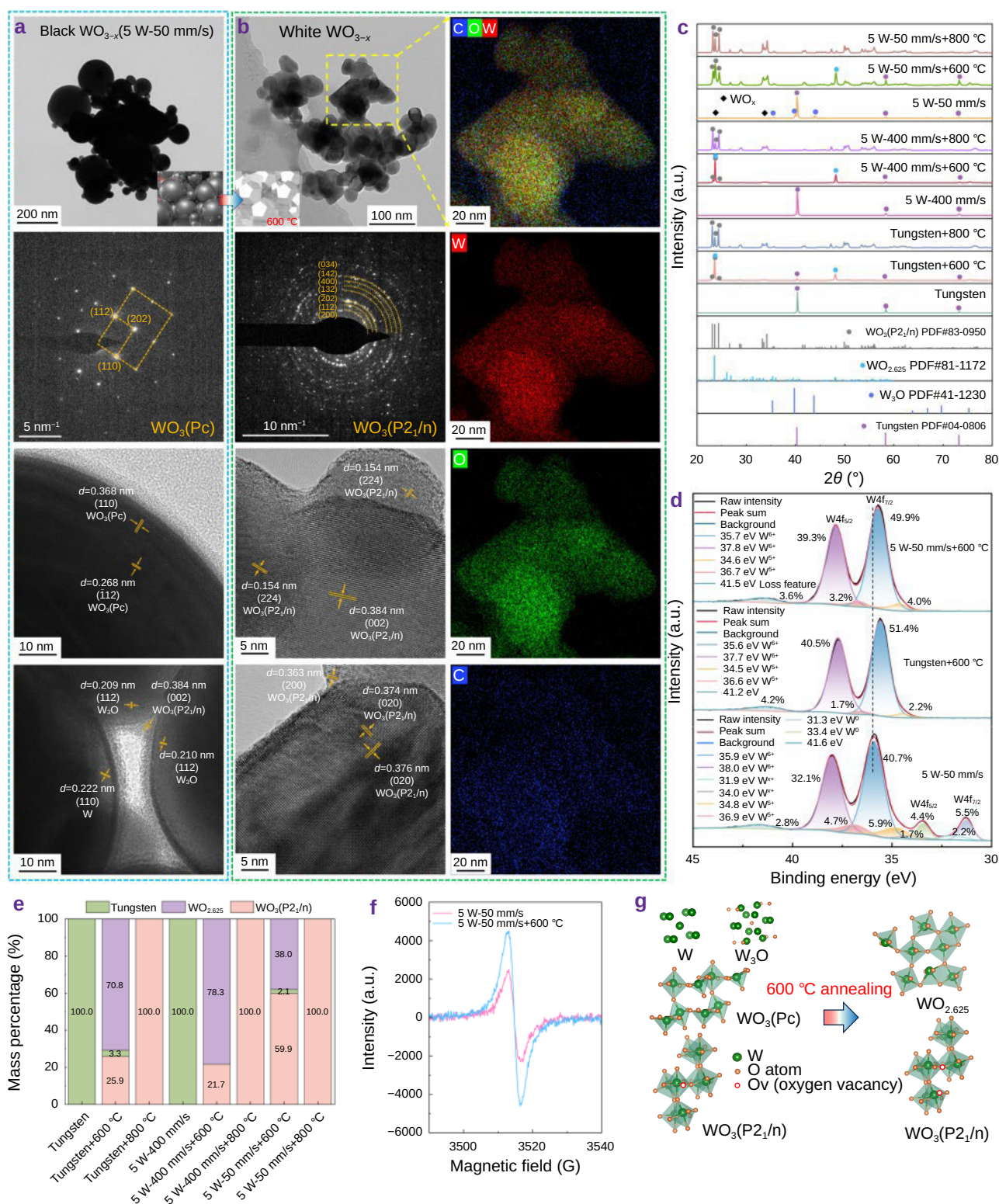


Fig. 2 | Characterization of WO_{3-x} nanostructures. (a, b) TEM, SAED, HRTEM, and EDS characterizations of black WO_{3-x} NPs (5 W-50 mm/s) and white WO_{3-x} (5 W-50 mm/s+600 °C). (c) XRD spectra of phase transitions of pristine W, black NPs, LIPSS (5 W-400 mm/s) and those obtained after 2h-annealing at 600/800 °C. (d) W4f and O2s XPS spectra of annealed W, black WO_{3-x} , and white WO_{3-x} . (e) Phase percentage quantification from XRD spectra. (f) EPR spectra of black and white WO_{3-x} . (g) Schematic of PT-to-RC black-to-white WO_{3-x} phase/structural evolution upon 600 °C annealing.

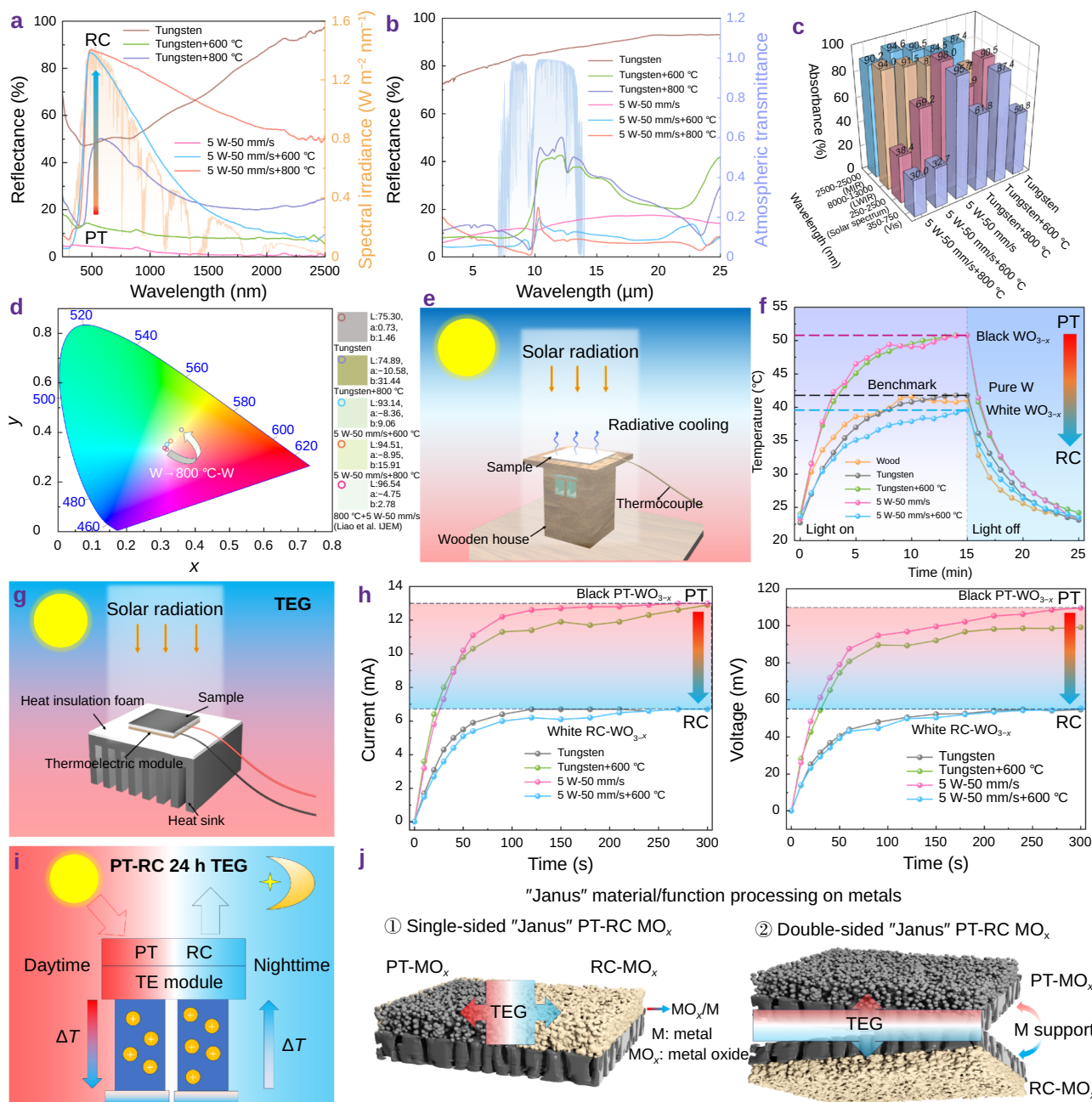


Fig. 3 | Optical, photothermal, and photothermoelectric properties. (a, b) UV-NIR and MIR spectra of pure tungsten, black WO_{3-x} NPs and their counterparts after 600/800 °C annealing. (c) Quantified absorbance in Vis, solar spectrum, LWIR, and MIR bands. (d) CIE 1931 chromaticity diagram showing different structural colors calculated from reflectance, compared with fs laser synthesized white WO_{3-x} aerogels³⁰. Curve arrow demonstrates broader color alteration upon 800 °C-annealing of pure W. (e) Schematic of the solar irradiation system for RC testing (1-sun intensity) on a wooden house. (f) Time-dependent temperatures profiles of the wood covered by pure tungsten, blue, black, and white WO_{3-x} samples. (g) Schematic of the daytime PT-TEG setup. The sample is located on a TEG module fixed on a heat sink. (h) I-V curves of TEGs using as-prepared samples. (i) Schematic of 24-hour PT-RC TEG application scenario using black and white WO_{3-x} . (j) Schematic of potential single/double-sided "Janus" PT-RC interfaces via fs laser ablation of refractory metals and subsequent thermal annealing treatment.

between white WO_{3-x} materials and the W substrate. Compared with the distinct color jump upon 800 °C annealing of pure W, coloration shifts a little after 600/800 °C annealing of fs laser synthesized black NPs. The band gap of white WO_{3-x} is ~ 2.86 eV, which is a little higher than the

2.70 eV bandgap of white WO_{3-x} nanofibers obtained by oxidative calcination³³. The blue WO_{3-x} (600 °C-W) exhibits absorbances of 87.4%, 90.5% and 74.8% in Vis, solar and LWIR bands, respectively. The high NIR absorbance is attributed to the oxygen-vacancy correlated with the local-

ized surface plasmon resonance (LSPR) effect⁴³.

Using the custom-built system shown in Fig. 3(e), we verified the PT/RC effects of black/white WO_{3-x} under 1-sun irradiation. The temperatures of the white- and black-WO_{3-x} samples are 2 °C lower and 9 °C higher, respectively, than that of the benchmark tungsten plate (Fig. 3(f)). This demonstrates that PT/RC-WO_{3-x} functionalized refractory metals can be effectively fabricated by fs laser in situ synthesis combined with simple thermal post-annealing at 600 °C. The temperature difference of ~11 °C between the black PT-WO_{3-x} and white RC-WO_{3-x} is sufficient for TEG applications⁴⁴. PT, RC, and PT-RC-integrated TEG has garnered significant attention for applications in distributed water desalination and energy harvesting^{45,46}. Using the TEG setup shown in Fig. 3(g), the current and voltage outputs for white, blue, black WO_{3-x}, and pure tungsten samples were measured (Fig. 3(h)). TEG performance relies on the temperature difference across the bottom and top interfaces of the thermoelectric device. Notably, black WO_{3-x} outperforms blue WO_{3-x} in PT-TEG application, yielding higher current and voltage outputs. This enhancement is attributed to the higher thickness of the black WO_{3-x} film, which allows for superior heat storage and thus higher temperatures upon light irradiation³⁵. The layer thickness of blue WO_{3-x} is approximately 6 μm thick³⁵, while the black WO_{3-x} NPs are formed solely by fs laser ablation of the pure W plate, without thermal annealing. The hierarchical NPs located on LIPSS effectively trap incident light to enhance the PT effect¹³. Furthermore, the oxygen vacancies within the structure contribute plasmonic “hotspot” properties, further boosting solar light absorption⁴⁷ and photothermal-electricity transition¹⁹.

Although the RC effect initially mitigates thermal accumulation, the TEG performances of white WO_{3-x} and pure tungsten samples become identical after 250 s. This indicates that heat yielded through solar absorbance is stored within the WO_{3-x} hierarchical structure. The thermal conductivity of tungsten is 170 W/(m·K), implying that heat can not be retained at the interface, which should rapidly dissipate into the ambient environment. In contrast, the thermal conductivity of WO_{3-x} is only 1–2 W/mK¹⁰, which is orders of magnitude lower. The identical TEG performances demonstrate that the RC effect of white WO_{3-x} effectively balance interfacial heat accumulation. The RC effect is expected to be more effective for TEG applications at night. Therefore, black/white WO_{3-x} pairs are promising candidates for Janus PT/RC-TEG applications, enabling continuous 24-hour electricity generation (Fig. 3(i)). This approach supersedes static, single-mode TEG systems that are limited to diurnal operation⁴⁸. Previously, fs laser fabricated black absorbers could only be combined with conventional SiO₂ and other RC prototyping methods for PT/RC-integrative TEG applications²². The processing paradigm presented here, when integrated with other established fs laser-based techniques^{29,30}, may pave new paths for developing horizon-

tal⁴⁹ and vertical⁵⁰ metal-supported oxide Janus thermal-regulation material/function matrices (Fig. 3(j)).

2.3 Single/double-sided PT/RC-integrated “Janus” processing and PT/RC-interface stability

To validate the potential “Janus material/functional” processing capacities, we performed single/double-sided integrated “white/black” processing on a single W-plate, as schematically illustrated in Fig. 4(a). First, PT-WO_{3-x} nanomaterials are prepared by fs laser ablation of W on one side and annealed at 600 °C to develop a white RC interface. Subsequently, the reverse side was processed by fs laser synthesis and in situ deposition of PT nanomaterials at 5 W and 50 mm/s. Despite the prior annealing, the interface remains unaffected by fs laser blackening, allowing for the convenient preparation of a Janus PT-RC black/white double-faced sample. Finally, the text “LASER JANUS” was patterned onto the white RC interface via selective laser scanning, realizing smart Janus PT-RC interfacial functionalization and information input (Fig. 4(b)).

The black pattern on the front exhibits high fidelity with no defect, while the completely black PT interface on the back, visualized through mirroring, was similarly defect-free. Consequently, the Janus PT-RC black/white double-faced structure integrated with two kinds of M-supported MO_x PT-RC Janus interfaces as demonstrated in Fig. 3(j), are successfully integrated. This achievement establishes an innovative “Janus” thermal-material/function processing paradigm for hard-to-process refractory metals like tungsten, which possess extremely high melting points. Compared to the PT/RC Janus film (RC-Al/SiO₂ with solar reflectivity of ~85% and IR emissivity ~95%; PT-CNTs with solar absorption of ~95%) which show temperature drop/increase of ~2 °C and ~7 °C⁸, the fs laser based RC/PT-WO_{3-x} interfaces delivered enhanced thermal regulation of ~2 °C reduction and ~9 °C elevation (Fig. 3(f)), confirming their practical applicability. Thermal stability and mechanical stability were assessed with the results shown in Fig. 4(c). From visual observation, the PT interfaces prove highly stable, withstanding 4 h thermal treatment at 100 °C/200 °C, vigorous human-hand shaking, 10 cycles of tape peeling, and 1 min of ultrasonic bath treatment in ethanol. Conversely, the RC interfaces only fail under ultrasonic bath, retaining integrity under all other tests. This vulnerability is attributed to the disintegration of porous NPs networks by high frequency (40 kHz) shaking in the liquid, removing particles/structures and revealing the underlying LIPSS and annealed-LIPSS structures (Fig. 4(d)). The resistance of NPs-network skeletons against mechanical stresses (shaking/peeling) and the high stability at 100 °C, facilitate their usage for common outdoor PT/RC applications^{51,52}, yet the pre-evaluation of interfacial adhesion, particularly under high-frequency ultrasonic stress condition, is as well essential for harsher-condition applications.

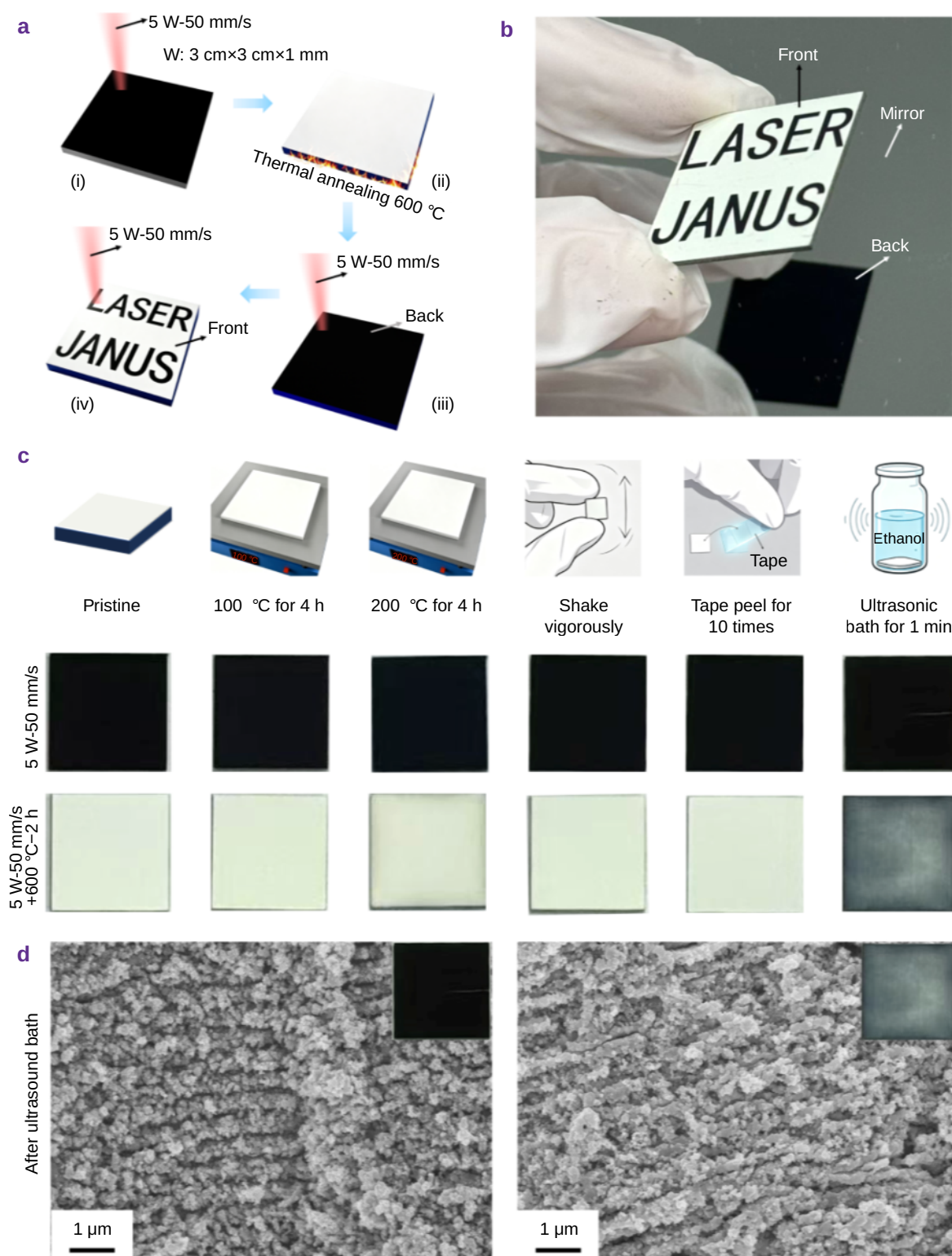


Fig. 4 | PT/RC-integrated "Janus" processing and thermal/mechanical interface stability. (a) Schematic of the Janus processing workflow on a W plate: (i) fs laser synthesis of black PT nanomaterials (5 W-50 mm/s); (ii) thermal annealing at 600 °C for RC whitening; (iii) fs laser PT manufacturing on the back side; and (iv) "LASER JANUS" patterning on the RC interface. (b) Optical image of the Janus sample, showing the PT "LASER JANUS" pattern on the RC white interface (top) and the mirror-reflected PT interface on the back side. (c) Thermal and mechanical stability tests. The RC (5 W-50 mm/s + 600 °C) and PT (5 W-50 mm/s) interfaces were subjected to thermal treatment (100 °C and 200 °C for 4 h) and mechanical stresses (vigorous shaking, 10-cycle tape peeling, and 1-min ultrasonic bath in ethanol). (d) SEM images of the surface morphologies of PT and RC samples after 1-min ultrasonic bath in ethanol.

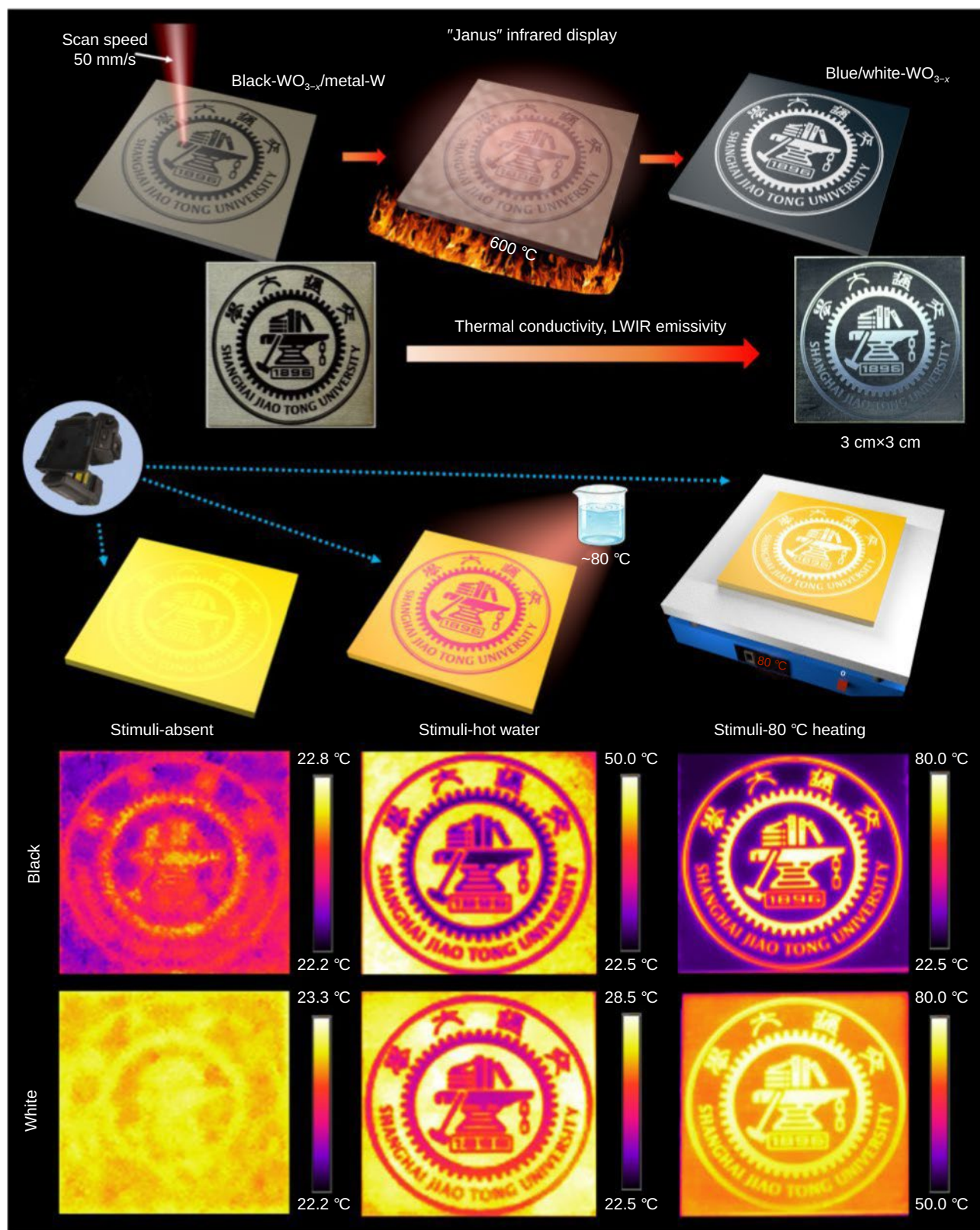


Fig. 5 | "Janus material/function" modulated IR display of "Shanghai Jiao Tong University" badge using black-WO_{3-x}/W and RC-white/PT-blue WO_{3-x} matrix. IR display is based on LWIR band camera. Stimuli-absent/present modes are comparatively investigated. External stimuli include hot water (~80 °C) IR reflection stimulus and 80 °C sample heating IR emission stimulus. The sample has a dimension of 3 cm × 3 cm. Schematic illustration of different stimuli scenarios is presented for a better understanding.

2.4 “Janus material/function” processing for display modulation

Figure 5 illustrates the fabrication of a “Shanghai Jiao Tong University” badge via fs laser-based W/black- WO_{3-x} Janus-material matrix patterning followed by white-RC/blue-PT- WO_{3-x} Janus functional transformation through 600 °C thermal annealing. The intricate details of black pattern remain clearly preserved after the transformation. Minor variations in lightness across different regions of the badge arises from subtle heterogeneities in the laser patterning and thermal annealing processes. Physically, pure W exhibits high thermal conductivity and high IR reflectivity, whereas black- WO_{3-x} demonstrates low thermal conductivity and high IR emissivity. Differently, the annealed blue and white WO_{3-x} interfaces possessing inverted solar responses and differential LWIR emissivities, collectively form a “Janus” optical/thermal modulator. In the absence of any external IR stimulus, the information remains IR cryptic and indistinct. However, under environmental IR stimuli such as exposure to hot water (~80 °C) or direct sample heating (80 °C), the system exhibits a modulated IR display response^{29,30}. According to the Stephan–Boltzmann law ($E = \sigma \epsilon T^4$, IR emissivity ϵ , Stephan–Boltzmann constant σ)⁵³, emitted IR radiation energy (E) scales significantly with temperature (T), enabling differential IR display in the heating mode. Due to the high thermal conductivity, heat dissipates rapidly from the pure W regions, whereas it accumulates in the black- WO_{3-x} pattern, generating a high-contrast IR image at larger temperature differentials. In contrast, heat accumulates uniformly in both the blue and white WO_{3-x} matrix regions; given their similar IR emissivities, these areas give rise to a warm, low-contrast IR display. The stark difference in IR reflectivity between W and black- WO_{3-x} produces a high-contrast image at a large temperature difference, while the little IR-reflectivity difference between blue/white WO_{3-x} leads to a relatively lower-contrast IR display. The switchable low-to-high contrast IR display is highly advantageous for developing environmentally responsive information encryption and decryption technologies.

2.5 Colorful “Janus material/function” solar thermal regulatory cooling

To verify the applicability of the proposed fs laser based “Janus material/function” processing paradigm to other transition metals (TMs) possessing gradient melting temperatures from > 3000 °C to 420 °C (Fig. 6(a)), black PT nano-materials are synthesized by fs laser ablation of Ta, Mo, Nb, and Ti at a laser power of 5 W and a scan speed of 200 mm/s, respectively. These PT interfaces are subsequently subjected to thermal annealing at 600 °C. Comprehensive UV-NIR and MIR spectra characterizations, along with quantifications of absorbance in Vis, solar, LWIR, and MIR bands, are presented in Fig. 6(b–d). The as-prepared Ta-,

Mo-, Nb-, and Ti-based PT interfaces exhibit comparable solar reflectivities in the range of 8%–9%, demonstrating the universality of this paradigm for developing PT absorbers regardless of the specific material or resultant transition metal oxide (TMOs) phase. Following 30 min of annealing, the black Ta/Mo/Nb-based PT interfaces undergo distinct color transitions to blue light, white, and green white, respectively, while their solar reflectivities increase to 24.6%, 45.7% and 52.2%, maintaining the LWIR emissivity of ~90%. In contrast, the Ti-based PT interface remains largely unchanged even after prolonged annealing for 2 h, exhibiting only minor reductions in solar reflectivity (1%) and LWIR emissivity (1%). This divergence in coloration or preservation upon 600 °C thermal annealing is intrinsic to the specific physicochemical properties of each TM/TMOs system^{19,35}.

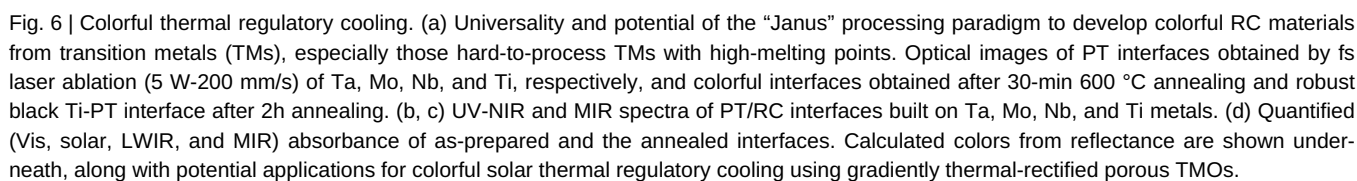
Conventional RC interfaces, typically exhibiting silver and white hues, face limitations in aesthetic integration in daily life and industrial applications, thereby driving increasing global interests in colorful RC materials⁵⁴. While coloration generally involves a slight compromise in RC performance due to altered visible light absorption⁵⁵, this trade-off opens new avenues for colorful solar thermal regulatory cooling⁵⁶. In this context, Ti-, Ta-, W-, Mo-, and Nb-based RC materials characterized by distinct colors (quantified by the CIELAB ($L^*a^*b^*$) color space), varying solar reflectivities, and high LWIR-emissivities, function as gradient thermal-cooling rectifiers (Fig. 6(d)). Given that solar spectrum comprises approximately 4% UV, 43% Vis, and 53% NIR light radiation⁵⁷, the thermal regulation mechanism under solar irradiation can be modeled by the following equation:

$$P_{\text{net}} = P_{\text{sun}}^{\text{UV}} + P_{\text{sun}}^{\text{Vis}} + P_{\text{sun}}^{\text{IR}} - P_{\text{cooling}}$$

Thermal rectification is primarily governed by absorption and reflection in the visible and infrared bands, as well as LWIR radiative cooling. However, high NIR absorbance remains a critical bottleneck that limits further enhancement of thermal rectification capacity (Fig. 6(d)). Oxygen vacancy engineering offers an effective strategy to modulate NIR absorbance in TMOs. This modification not only influences material coloration⁵⁸ but also induces the LSPR effect to mitigate heat loss associated with radiative cooling and enhance interfacial photothermoelectric conversion¹⁹.

2.6 Pros and cons of fs laser-based thermal regulation “Janus material/function” processing paradigm

To elucidate the pros and cons of fs laser-based thermal regulation “Janus material/function” processing paradigm (encompassing techniques, products and PT/RC properties), we benchmark our results against those derived from alternative methodologies, as summarized in Table 1. Conventionally, PT and RC interfaces are developed in isolation, inherently disenabling the PT-to-RC transition. This integration is the primary breakthrough of the presented work.



A previous report on the PT-to-RC transition utilized black silicon slag (SIS) exhibiting intrinsic solar reflectivity of 11% and LWIR-emissivity of 90.8%⁵⁹. Annealing in a muffle furnace at 1100 °C for 24 h enhanced LWIR-emissivity to 96.2% and solar reflectivity to 24%. The incorporation of Al to modify the crystal structure only marginally increased the solar reflectivity to 32%. By comparison, our proposed method is superiorly energy efficient, enables information encoding, and demonstrates broad applicability across different metallic substrates.

SiO₂ is the most common RC material. However, aggregates of 300 nm SiO₂-NPs merely achieve a solar reflectivity of 55.5% and an LWIR emissivity of 78%⁶⁰, which is close to the performance of NbO_x-aggregates after the PT-to-RC transition. Since SiO₂ is transparent, it is often used as a filler in polymer matrices to gain high solar reflectivity and high LWIR emissivity. To prevent light emission from the substrate, coating of a highly reflective metal is required, such as SiO₂@PDMS/Al-support possessing 85% solar reflectivity and 95% LWIR emissivity⁸. Adding SiO₂ NPs to printable polymers allows for creation of 3D-RC structures⁶¹. Colorful RC films are often made by coating commercial paints on polymer composite⁶² or by mixing color pigments or phosphor dyes with inorganic reflective/emissive powders in a polymer matrix^{63,64}. Pre-designed masks allow colorful patterning in self-stratified photonic RC composites⁶⁵. However, since polymers degrade under UV irradiation, purely inorganic RC structures are more valuable for their excellent stability in harsh conditions like outer space. Fs laser has been used to synthesize different TMOs-NPs aerogels with a higher solar reflectivity of 70.6% and higher LWIR emissivity up to 99.6%³⁰. These aerogels are dispersible in liquids, and the alternatives directly synthesized by laser ablation in liquids⁶⁶ can both further improve RC performance.

RC metasurfaces and metamaterials, integrated with emissive micro/nanostructures, are garnering increasing worldwide attention due to their precise regulation capacities of interfacial optical and thermal properties, and their reduced reliance on degradable polymer matrices⁶⁷. However, structuring bulk SiO₂ substrates typically relies on time-consuming expensive equipment, such as reactive ion etching⁶⁸. To construct a high-performance colorful metasurface emitter, both the colored emitting layer and the substrate reflector must be meticulously optimized. For instance, Xia and Fan developed a colored radiative cooler based on a metal-insulator-metal (MIM) photonic reflector⁶⁹. The upper cooling layer is composed of SiO₂ cone units (fabricated via lithography and wet etching using a sacrificial layer), TiO₂ layer (deposited via magnetron sputtering), and a spin-coated polydimethylsiloxane (PDMS) layer. Although geometric parameters allow for emissivity control, the fabrication process is highly complex, requiring the coordination of multiple materials and specialized equipment⁶⁹. In contrast, laser patterning offers multi-

dimensional processing capacities such as mask fabrication⁷⁰, sculpturing⁷¹, and high-throughput processing of transparent materials^{72,73}. Leveraging techniques such as 3D multi-scanning and dense-stacking^{74,75} can dramatically increase information density, facilitating large-scale secure information storage. Furthermore, the in situ fs laser synthesized TMOs-NPs is directly transferable to transparent substrates. When combined with machine learning customization⁷⁶, this approach paves the way for advanced 3D multifunctional fs laser-based Janus manufacturing, with inspirations drawn from natural structures^{77–80}. In addition, TMOs such as Nb₂O₅⁸¹, WO_{3-x}⁸² and MoO₃⁸³ exhibit photochromic properties, enabling reversible modulation of their color appearance through UV irradiation and thermal treatment in air, which can well circumvent the accurate optical design and delicate material optimization required for reversible display in the system of Tamm plasmonic photonic crystals⁸⁴.

Refractory metals offer a robust, integrated substrate system for tunable PT/RC regulation, exhibiting superior environmental stability compared to polymer matrices. Furthermore, they eliminate the need of additional metal coatings, polymer blending, or complex prototyping procedures. The integration of RC-active porous TMOs NPs-networks onto these metallic supports is governed by surface morphology and the associated oxide crystallization (Fig. 3(j)). In this context, interfacial structures such as LIPSS play a critical role in determining the adhesion strength. The formation mechanism of LIPSS on metals involves transient melting⁸⁵, laser-induced oxidation⁸⁶, and excitation of electromagnetically coupled surface plasmon polaritons (SPPs)⁸⁷. LIPSS and deep-subwavelength structures can emerge upon laser-matter interaction; however, their formation is competitive and highly dependent on the localized effective laser fluence⁸⁸. By tuning laser spatial polarization, the orientation of LIPSS can be modulated^{89,90}, thereby tailoring the specific surface area and corresponding connection sites. The versatility of these interface structures, combined with the ability to adjust the density of deposited TMOs-NPs, provides a significant flexibility for optimizing interfacial thermal performance.

Beyond the prevalent PT-to-RC transition, PT-resistant interfaces have also been developed, as exemplified by the black Ti-absorbers merely undergoing 1% and 1% alteration of solar reflectivity and LWIR-emissivity, respectively, after 2 h-600 °C annealing. This robustness makes them good candidates to be used as thermal emitters, which may expand the scope of thermal regulation capacities, complementing laser grooved metallic emitters (emissivity > 96%) that operate steadily at temperatures exceeding 1500 °C⁹¹ and metallic blackbody with > 98% absorption across the 0.2–25 μm spectrum²⁵. The in situ fs laser synthesized PT-WO_{3-x} NPs-aggregate has been employed for black-color information encryption via multiple environmental stimuli (e.g., human hand, hot/cold water, ice, and sample

Table 1 | Comparison of as-prepared colorful RC materials with reported RC materials.

Technique	Substrate (composition)	Solar reflectivity	LWIR emissivity	PT-RC transition	Colorful	Patternability	Ref
Fs laser synthesis + thermal annealing	Porous WO _x , TaO _x , MoO _x , NbO _x , TiO _x NPs networks	30.8% 24.6% 45.7% 52.2% 9%	91.5% 71.8% 76.1% 88.8% 88.9%	Yes	Yes	Yes	This work
Fs laser synthesis	SiO ₂ , ZrO ₂ , Al ₂ O ₃ , TiO ₂ , WO ₃ NPs aerogels	69.4% 70.6% 56.7% 19.6% 30.4%	99.6% 96.2% 99.3% 98.5% 95.4%	No	Yes	Yes	Ref. ³⁰
High temperature annealing (Al assisted)	Black silicon slag	11%, 24%, 32%	90.8%, 96.2%, 95.77%	Yes	No	No	Ref. ⁵⁹
Commercial	SiO ₂ NPs aggregation (100 nm, irregular)	55.5%	78%	No	No	No	Ref. ⁶⁰
Solution deposition	SiO ₂ @PDMS/Al-support	85%	95%	No	No	No	Ref. ⁸
3D printing	CNFs; CNFs + SiO ₂	71.95 %; 94.18 %	89.73 %; 96.09 %	No	No	Yes	Ref. ⁶¹
Solution drop casting on glass substrate	h-BN NPs + color pigment dispersed in PU polymer matrix	86%–89%, 86%–89%, 77%–79%, 97%–99%	86%, 86%, 85%, 89%	No	Yes	No	Ref. ⁶³
Self-stratifying hierarchical structuring	Top photonic CNC layer + bottom BN-rich layer + pre-patterned templates	96.9%	91.5%	No	Yes	Yes	Ref. ⁶⁵
Paint coatings	Porous P(VdF-HFP) or TiO ₂ /polymer composite + commercial paints	blue 40%; red 61%; yellow 72%	all 95%	No	Yes	No	Ref. ⁶²
Paint coatings	Polyacrylic resin + TiO ₂ /BaSO ₄ powders + phosphor dyes	92.8%; 93.7%; 92.3%; 90.8%	94%; 93%; 93%; 92%	No	Yes	No	Ref. ⁶⁴
lithography + reactive ion etching + magnetron sputtering + spin coating	Top emitter composed of SiO ₂ cone units, TiO ₂ layer and PDMS layer; lower reflector consists of Ag/SiO ₂ elliptical cylinders	–	96.62 %	No	Yes	Yes	Ref. ⁶⁹

Note: polyurethane (PU), cellulose nanofibers (CNFs), poly(vinylidene fluoride-co-hexafluoropropene) [P(VdF-HFP)]

heating/cooling) IR decryption²⁹. It is deemed that the high thermal resistance of TiO_x-PT interfaces could raise the thermal threshold for information decryption and IR decryption up to 600 °C. Under these conditions, refractory metals such as W, Ti, and Ti-alloy (e.g., Ti6Al4V) undergo oxidation with distinct color changes¹⁹. Additionally, the room-temperature bandgaps of anatase and rutile TiO₂ are 3.24 and 3.16 eV, respectively, which significantly decrease at 600 °C⁹², enhancing their suitability for PT catalysis applications.

3 Conclusion

In this work, we validate the “Janus material/function” matrix processing paradigm by simple fs laser ablative synthesis of black TMOs NPs-aggregates or selective in situ patterning, followed by thermal annealing at 600 °C. The PT-black WO_{3-x} is directly converted into white RC-WO_{3-x} without compromising spatial resolution or inducing significant particle growth. Under 1-sun solar irradiation, the PT/RC-WO_{3-x} interface achieves a temperature increase of

9 °C and a decrease of 2 °C relative to pristine W, demonstrating superior thermal regulation capacities compared to the bare substrate. The distinct Janus functions of the W/black-WO_{3-x} interface, characterized by contrasting thermal conductivity and IR-reflectivity/emissivity, enable high-contrast IR displays under external thermal stimuli of 80 °C hot water approaching and sample heating. In contrast, the annealed white/blue-WO_{3-x} phases, which exhibit similar thermal conductivities and moderate differences in IR/solar optical properties, result in low-contrast IR displays. The significant differential thermal modulation underscores the efficacy of the proposed fs laser based processing paradigm for Janus thermal regulation from the material/function matrix perspective. Furthermore, extending this technique to other TMs such as Ta, Mo, and Nb, demonstrates its universality in manufacturing PT-TMOs and colorful PT-to-RC transitions. While black PT-TiO_x interface exhibits exceptional resistance to thermal oxidation, highlighting the versatility of this approach.

4 Materials and methods

4.1 Fs laser ablation of W and thermal annealing

Pure tungsten plates were purchased from Qinghe Lisheng Metal Material Co., Ltd. (China) with dimensions of 10 mm × 10 mm × 1 mm and 30 mm × 30 mm × 1 mm. The purchased Ta, Mo, Nb, and Ti plates were in the dimension of 30 mm × 30 mm × 1 mm. A femtosecond (fs) laser system (Trumpf, Trumicro 2030, Germany) with a pulse duration, wavelength, and repetition rate of 400 fs, 1030 nm, and 400 kHz, respectively, was used to ablate tungsten plates in an air atmosphere at focus. The tungsten plates were ablated at laser powers of 5 and 15 W and scan speeds of 400 and 50 mm/s, using a galvanometer, to prepare different structures. Throughout experiments, line-by-line scanning method was adopted, with the scan interval between adjacent lines set at 3 μm. Thermal annealing of tungsten plates and as-ablated structures was then performed in a muffle furnace (Nabertherm GmbH, L5/13/P330, Germany) at 600 °C or 800 °C for 2 h to investigate the impact of temperature on the resultant microstructure morphology and composition. To simplify the description, samples are defined by the sequence of laser power, scan speed and annealing temperature. For example, the 5 W-50 mm/s sample corresponds to the tungsten plate ablated by a fs laser at a power of 5 W and a scan speed of 50 mm/s, while the 5 W-50 mm/s+600 °C sample corresponds to the 5 W-50 mm/s sample subjected to post thermal annealing at 600 °C for 2 hours.

4.2 Optimal annealing condition screening

A series of experiments was performed on the 5W-50mm/s samples to further investigate the effects of the annealing temperature and duration on the changes in structural colors. The samples were annealed in a muffle furnace at

400 °C for durations of 5, 15, 30 min, 1 h, and 2 h, and at 600 °C for 1, 5, 15, 30 min, 1 h, and 2 h. In addition, comparative experiments were conducted by annealing samples at 300, 400, 500, and 700 °C for 2 h.

4.3 “Shanghai Jiao Tong University” badge patterning

Black patterning of “Shanghai Jiao Tong University” badge was obtained through fs laser scan at the laser power of 5 W and scan speed of 50 mm/s on a tungsten plate. The black badge and metallic color background turned to white and dark blue, respectively, after annealing at 600 °C for 2 h.

4.4 Universality of PT-interface processing on other metals and colorful RC function transition

Colorful transition-metal oxides (TMOs) were investigated by fs laser ablation of Ta, Mo, Nb, and Ti targets and subsequent thermal annealing. Fs laser ablation was performed at a power of 5 W and a scan speed of 200 mm/s (other parameters are in line with those of fs laser ablation of W) to obtain black PT nanomaterials. Line-by-line scanning method was adopted, with the scan interval of 3 μm. Then the black PT nanomaterials located on the metal support were thermally annealed at 600 °C. The annealing period is 30 min for PT-Ta/Mo/Nb samples and 2 h for PT-Ti sample.

4.5 Sample characterizations

Surface morphology and its elemental distributions were characterized by scanning electron microscopy (SEM; Sigma 500, Zeiss, Germany) equipped with energy-dispersive X-ray spectroscopy (EDS). The morphology, phase and elemental distribution of NPs' aggregates were characterized by transmission electron microscopy (TEM; Talos F200X G2, Thermo Scientific, United States) equipped with selected area electron diffraction (SAED), and energy dispersive X-ray spectroscopy (EDS). The phase composition was determined by X-ray diffraction (XRD; Mini Flex 600, Rigaku, Japan, using Cu Kα radiation, λ = 0.15406 nm). Raman spectra were obtained using a 532 nm laser confocal Raman micro-spectroscopy (Raman; Renishaw inVia Qontor, Renishaw, United Kingdom). Compositions and electron structures were characterized by X-ray photoelectron spectroscopy (XPS; AXIS UltraDLD, Kratos, Japan). And the presence of unpaired electrons (e.g., oxygen vacancies) was confirmed by Electron spin resonance spectra (ESR; Bruker A300, Bruker, Germany). The reflectance spectra were measured by the UV-Vis-NIR spectrophotometer (Lambda 950, PerkinElmer, Inc., United States) and Fourier transform IR spectroscopy (FTIR) (Nicolet 6700, Thermo Fisher, United States). The IR images of the “Shanghai Jiao Tong University” paintings were recorded by an infrared thermal imaging device (T630, FLIR, United States) at room temperature. Placing a beaker containing 80 °C hot water close to

the sample and 80 °C-heating of the sample for reflection/emission stimuli-responsive differential IR display, respectively.

4.6 Radiative cooling, photothermal evaluation and photothermoelectric generation test

The self-built solar irradiation system was adopted to evaluate the photothermal and radiative cooling effects of the 5 W-50 mm/s, 5 W-50 mm/s+600 °C, Tungsten+600 °C and pure tungsten samples by attaching them to the roof of a wooden house. A wooden piece was placed on the roof for a comparative study. A thermocouple put underneath the tested sample is used to record every-minute temperature change upon light irradiation for 15 min. A self-built solar irradiation system was designed to evaluate the photothermoelectric conversion ability of samples by attaching them to the upper side of the thermoelectric generator. The Xenon lamp (Xenon lamp, MC-PF300-UV, Merry Change, China) with the light intensity of 100 mW/cm² (one sun) was adopted to irradiate the tested samples for radiative cooling, photothermal, and photothermoelectric generation testing. The yielded voltages and currents were measured by a multimeter.

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Competing interests

The authors declare no competing financial interests.



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