

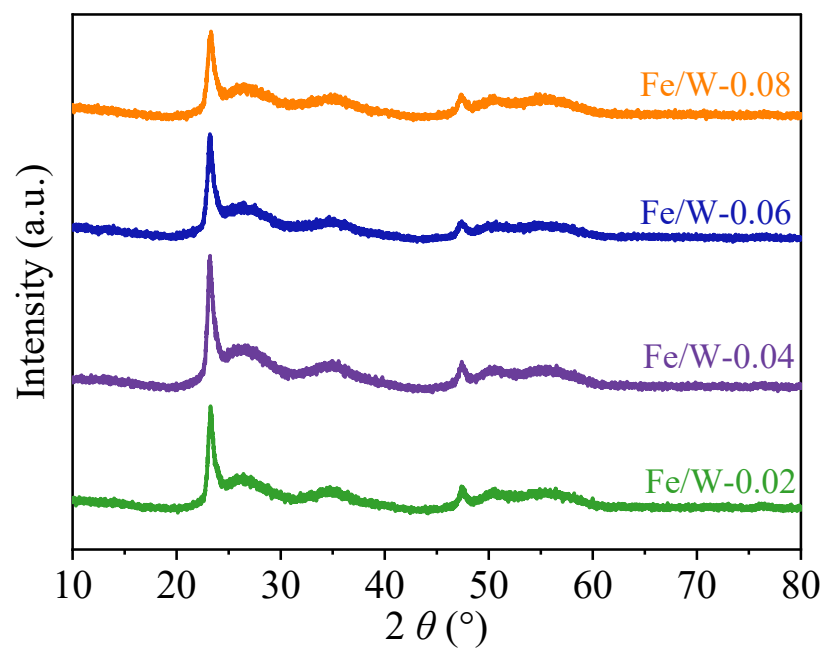
## **Transparent photochromic Fe-doped $W_{18}O_{49}$ films with ultrahigh solar energy modulation for smart windows**

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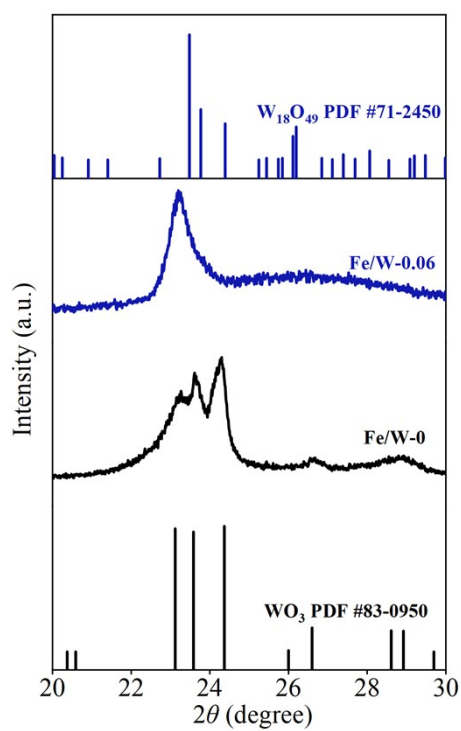
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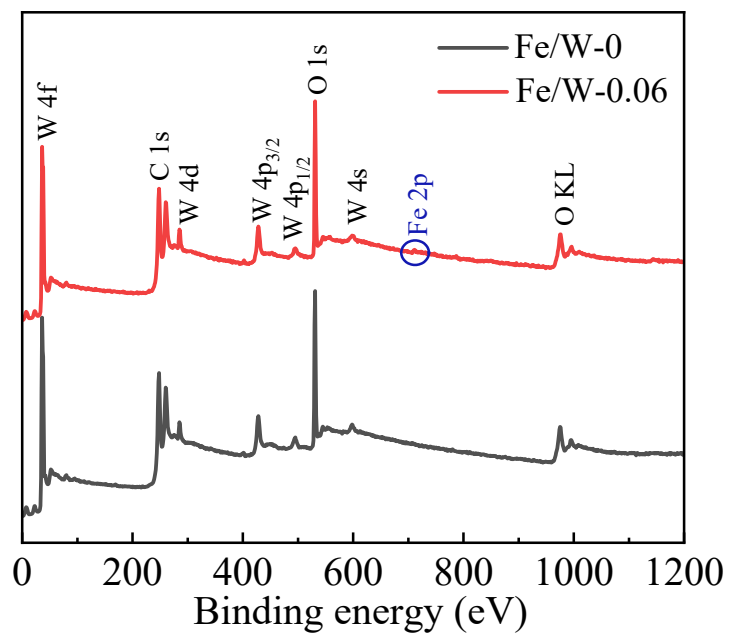
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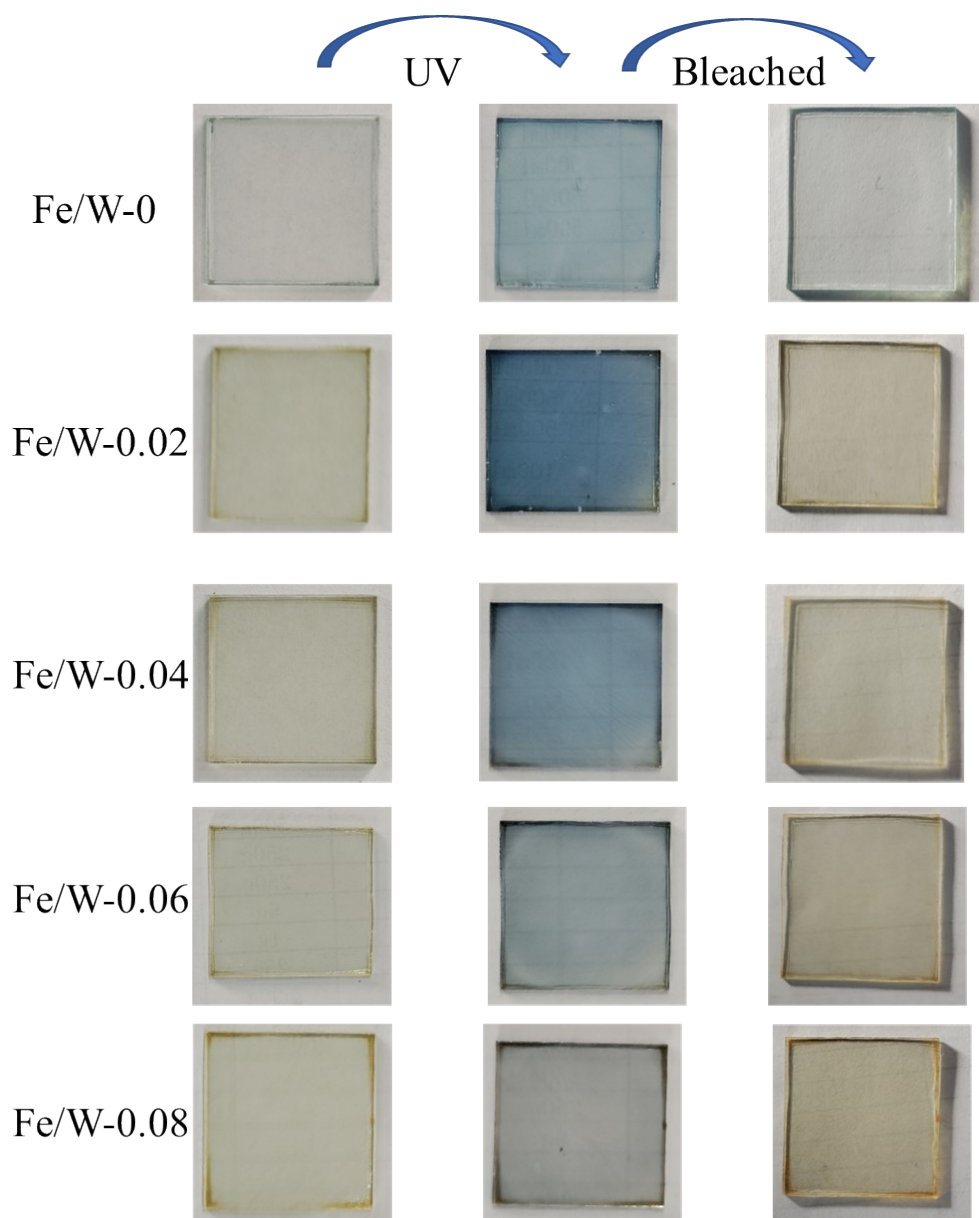
**Fig. S1** XRD patterns of Fe/W-0.02, Fe/W-0.04, Fe/W-0.06 and Fe/W-0.08 nanoparticles.



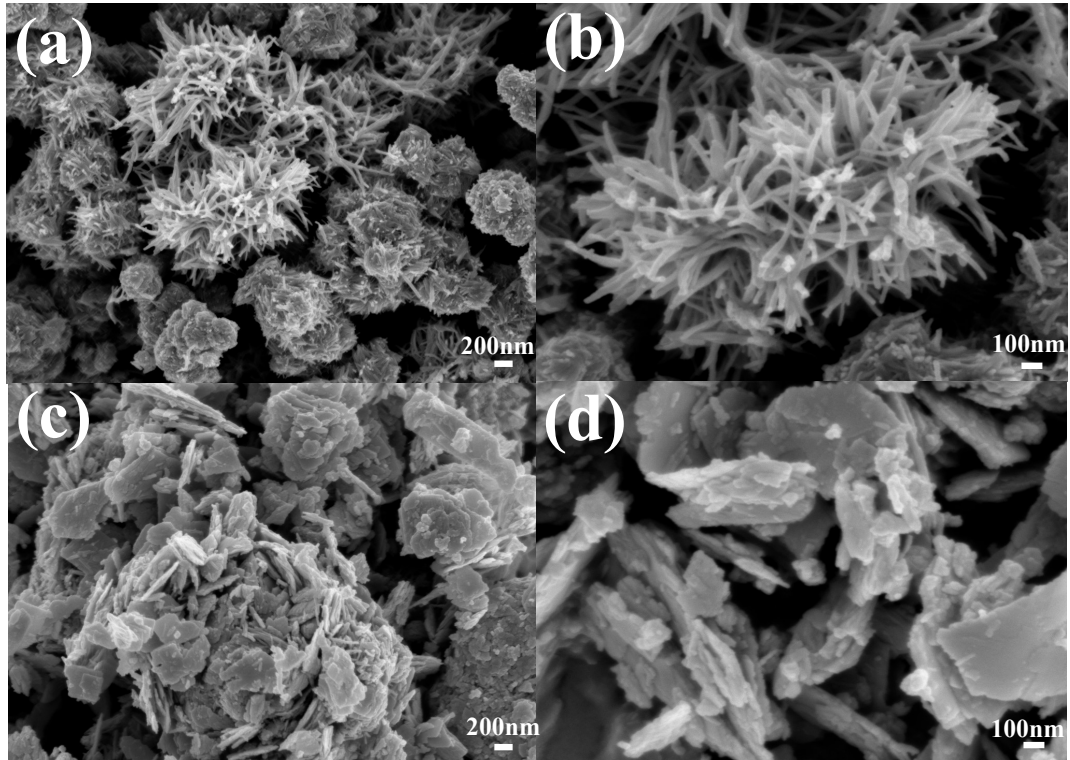
**Fig. S2** XRD patterns of Fe/W-0.06 and Fe/W-0 nanoparticles.



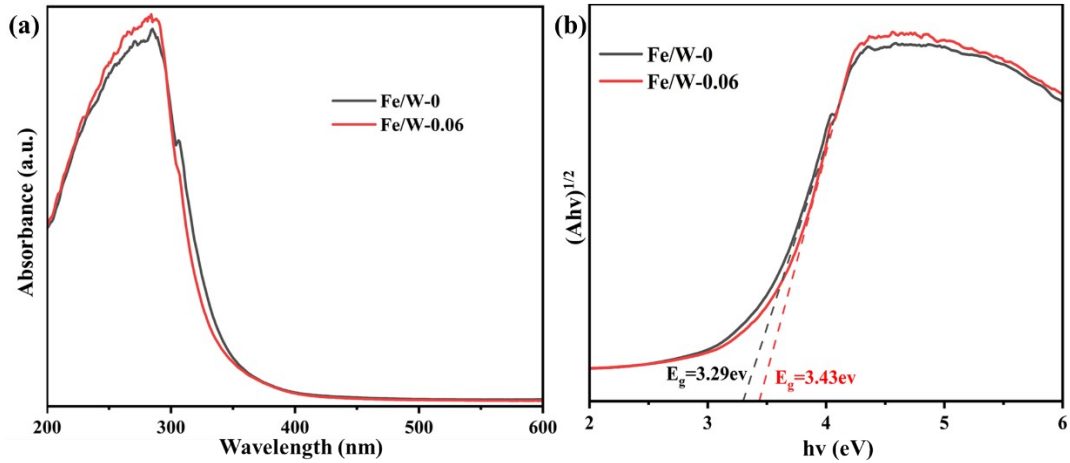
**Fig. S3** Survey XPS spectra of Fe/W-0 and Fe/W-0.06 nanoparticles.



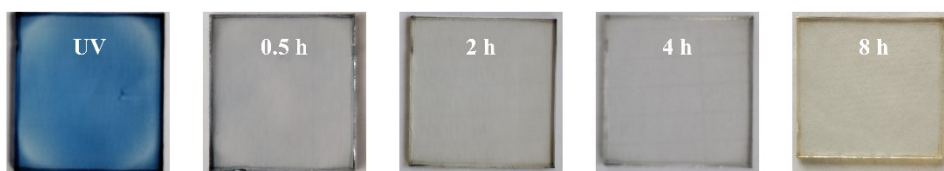
**Fig. S4** The photographs of Fe/W-0~0.08 films from bleached to colored to bleached state.



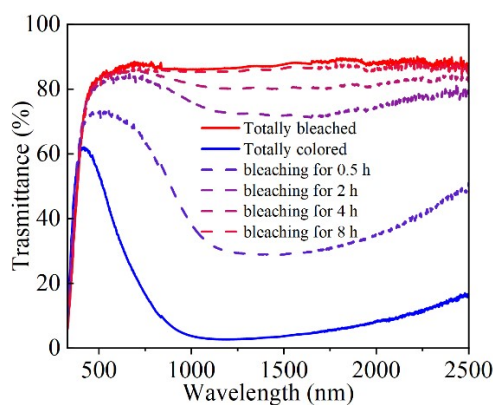
**Fig. S5** SEM image (a) and partial enlargement (b) of Fe/W-0 particles. SEM image (c) and partial enlargement (d) of Fe/W-0.06 particles.



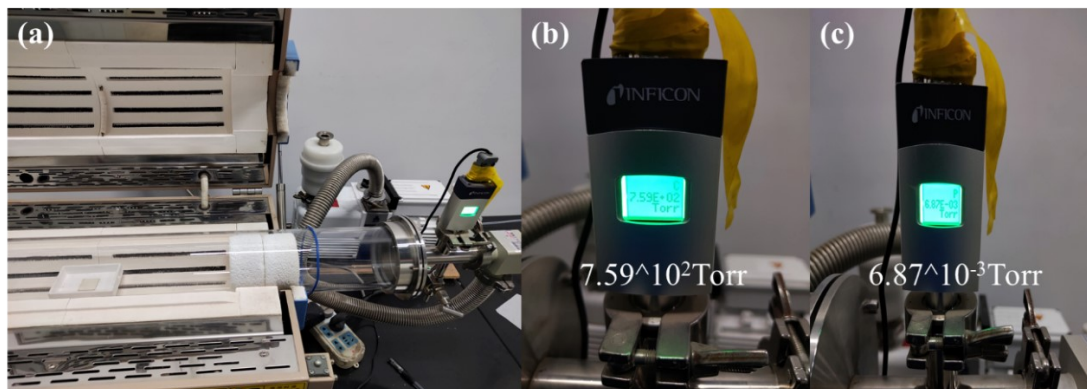
**Fig. S6** (a) UV-Vis absorption spectra of Fe/W-0 and Fe/W-0.06 particles, (b) the corresponding optical band gap. The optical bandgap  $E_g$  was derived from equation  $(\alpha hv)^m = A(hv - E_g)$ , where the exponent  $m$  here was identified as  $1/2$  for indirect-allowed optical transition. By linear fitting the plot of  $(\alpha hv)^{1/2}$  depend on  $hv$ ,  $E_g$  was obtained from the intercept of fitted line with x axis.



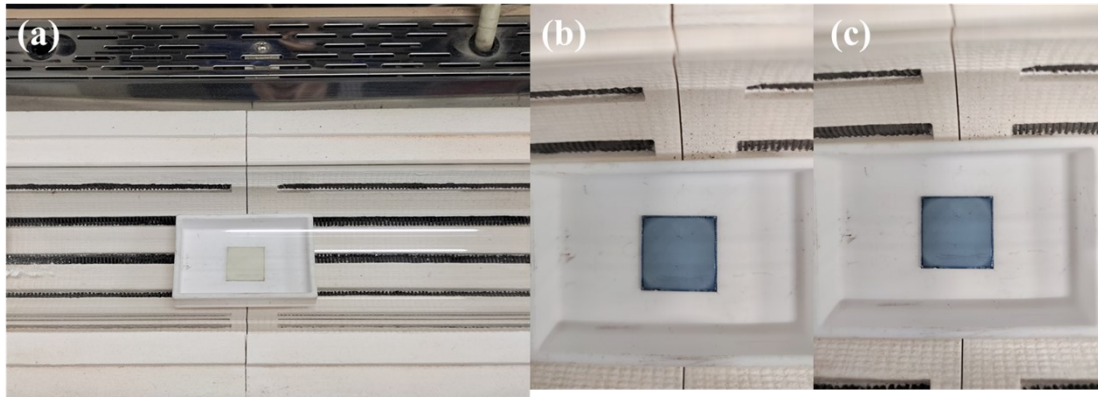
**Fig. S7** Photographs of Fe/W-0.06 bilayer film during the bleaching process at 15 °C in the dark.



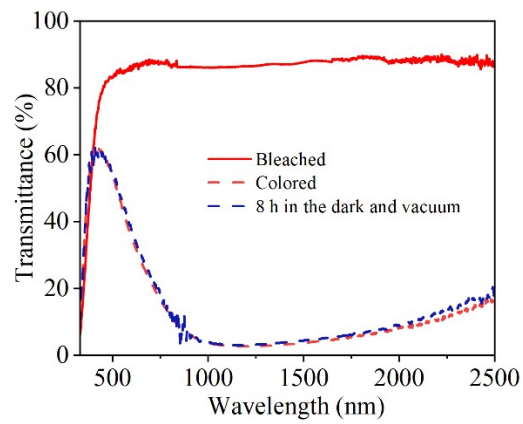
**Fig. S8** Transmittance spectra of Fe/W-0.06 bilayer film during the bleaching process at 15 °C in the dark.



**Fig. S9** (a) Photograph of the experimental set up. The pressure in the tube furnace (b) before and (c) after evacuation (the coloring and bleaching process were taken under this condition).



**Fig. S10** Photographs of Fe/W-0.06 bilayer films: (a) totally bleached, (b) totally colored and (c) the colored film after leaving in a vacuum tube furnace at room temperature in the dark for 8 h.



**Fig. S11** Transmittance spectra of Fe/W-0.06 bilayer film in the totally bleached and colored state, and the totally colored film after leaving in a vacuum tube furnace at room temperature in the dark for 8 h.

**Table S1** Optical properties of films with different doping Fe<sup>3+</sup> concentration.

Fe/W (at.%)	$T_{\text{sol}}$ (%)		$T_{\text{lum}}$ (%)		$\Delta T_{\text{sol}}$ (%)
	Bleached state	Colored state	Bleached state	Colored state	
0	83.0	45.5	87.3	67.8	37.5
2	84.4	35.6	88.0	46.7	48.8
4	85.1	41.1	87.5	57.0	44.0
6	84.3	42.6	87.2	58.3	41.8
8	84.9	53.6	87.2	69.6	31.3
6 (bilayer)	79.9	19.7	82.9	34.9	60.2

**Table S2** The transmittance of films with different doping Fe<sup>3+</sup> concentrations at 1200 nm during the bleaching process.

Fe/W (at.%)	Transmittance (%)					
	Colored state	B-5min	B-10min	B-20min	B-30min	Initial state
0	17.95	42.46	50.43	62.26	67.09	81.41
2	20.83	48.85	60.29	67.80	71.43	82.81
4	20.13	66.07	74.70	79.98	80.44	85.49
6	21.74	75.39	81.51	84.16	84.88	85.56
8	30.59	82.25	83.89	84.76	84.85	86.04



**Table S3** Comparison between the Fe/W-0.06 bilayer film and some best reported photochromic materials in terms of photochromic performance.

Materials	Irradiation conditions	Coloring process	Bleaching process	$\Delta T$	Ref.
WO <sub>3</sub> /PVA	365 nm, 8 W	5 min	Air/dark, 3 d	~30%	1
WO <sub>3-x</sub> /Ta <sub>2</sub> O <sub>5</sub>	365 nm, 8 W	10 s	–	28% at 1200 nm	2
Zn/W-0.04	365 nm, 5 W	<1 min	–	47.18%	3
WO <sub>3</sub> , TiO <sub>2</sub> and SiO <sub>2</sub>	15 min, sunlight,	-	air/dark, ~2 days	41% ( $T_{vis}$ )	4
Cu-WO <sub>3</sub>	90 min, 365 nm	-	air/dark, ~3h	-	5
Fe/W-0.06	365 nm, 5 W	<1 min	<20 min at 80 °C	60%	This work

## References

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- 4 M. Hočevár and U. Opara Krašovec, *Sol. Energy Mater. Sol. Cells*, 2018, **186**, 111–114.
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