

Kirkendall effect induced ultrafine VOOH nanoparticles and its transformation into VO₂(M) for energy-efficient smart window

Liangfei Wu,^{a†} Antonio Teng,^{b†} Ming Li,^{*ac} Liang Li,^{ac} Zhulin Huang,^{ac} Xinyang Li,^a Jie Yu,^a Sichao Xu,^a Fengxia Zou,^{ac} Andy Zou,^d Jinghui Zhang,^{e,f} Tao Jiang,^{e,f} Ye Xin,^g Xiaoye Hu,^{*ac} Guanghai Li^{*ac}

^aKey Laboratory of Materials Physics, Anhui Key Laboratory of Nanomaterials and Nanotechnology, Institute of Solid State Physics, Hefei Institutes of Physical Science, Chinese Academy of Sciences, Hefei 230031, P. R. China

^bContiTech ChinaRubber & Plastics Technology Ltd., Changshu 215500 P. R. China

^cUniversity of Science and Technology of China, Hefei 230026, P. R. China

^dBenecke Changshun Auto Trim Co., Ltd., Zhangjiagang 215632 P. R. China

^e Key Laboratory of Atmospheric Optics, Anhui Institute of Optics and Fine Mechanics, Hefei Institutes of Physical Science, Chinese Academy of Sciences, Hefei 230031, P. R. China

^f Advanced Laser Technology Laboratory of Anhui Province, Hefei 230037, P. R. China

^gNaval Research Institute, Beijing 102442, P. R. China

[†] The authors have equally contributed to this work.

*Corresponding author.

E-mail: liming@issp.ac.cn (M. Li), hxy821982@issp.ac.cn, ghli@issp.ac.cn

gradient		specific values for the constant parameters		
		Hydrothermal temperature	Reaction time	The volume of HCOOH
Hydrothermal temperature	80 °C	120 °C	6 h	1 ml
	120 °C			
	160 °C			
	200 °C			
Reaction time	1.5 h	120 °C	6 h	1 ml
	6 h			
The volume of HCOOH	0 ml	160 °C	6 h	1 ml
	0.5 ml			
	1 ml			
	1.5 ml			
	2 ml			

Fig. S1. The synthesis parameter for different gradient experiment.

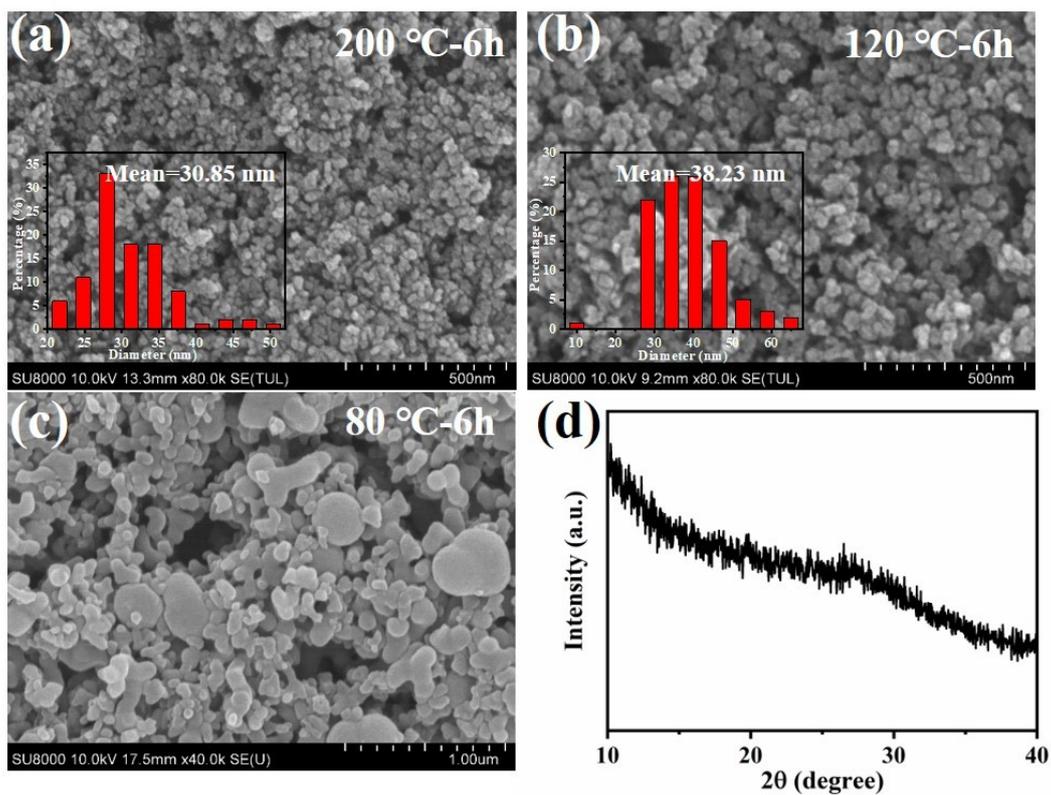


Fig. S2. The SEM images of VOOH nanoparticles prepared with different hydrothermal temperature: (a) 200 °C and (b) 120 °C. The SEM image and XRD pattern of hydrothermal product at temperature of 80 °C. (inset: histogram of size distribution images of the VOOH NPs).

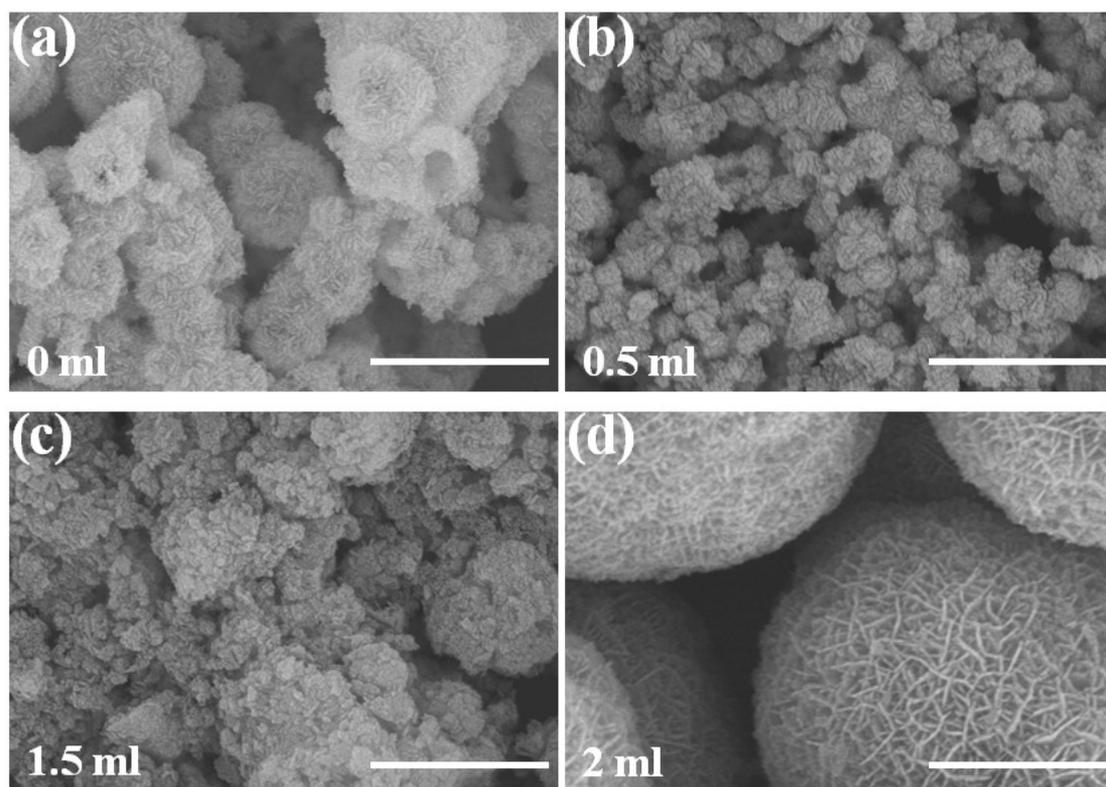


Fig. S3. The SEM images of hydrothermal products with different volume of HCOOH: (a) 0 ml, (b) 0.5 ml, (c) 1.5 ml and (d) 2 ml. (Scale bar: 1 μm)

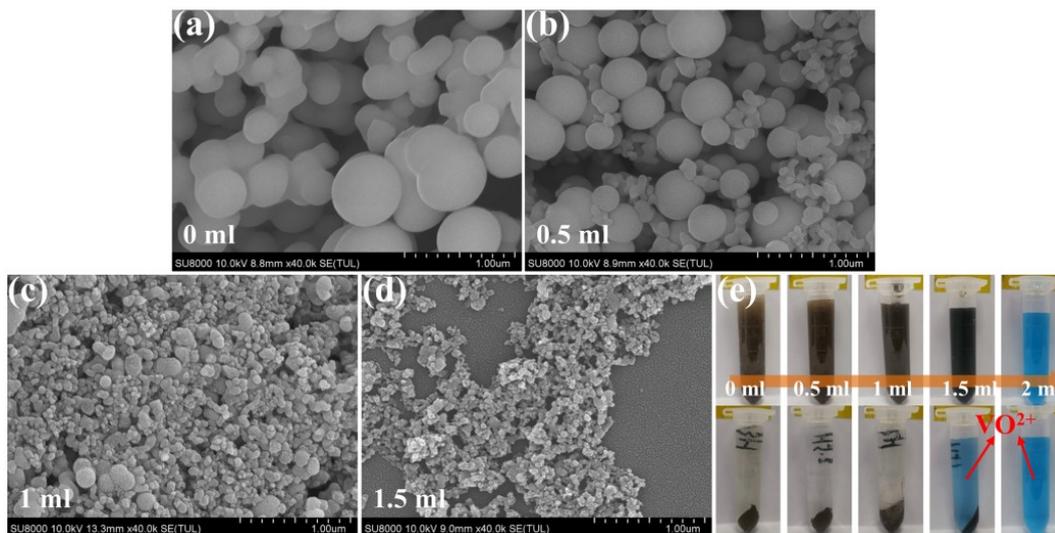


Fig. S4. The SEM images of hydrothermal precursors with different volume of HCOOH: (a) 0 ml, (b) 0.5 ml, (c) 1 ml and (d) 1.5 ml. (e) Digital images of hydrothermal precursors before (top) and after centrifugation (down).

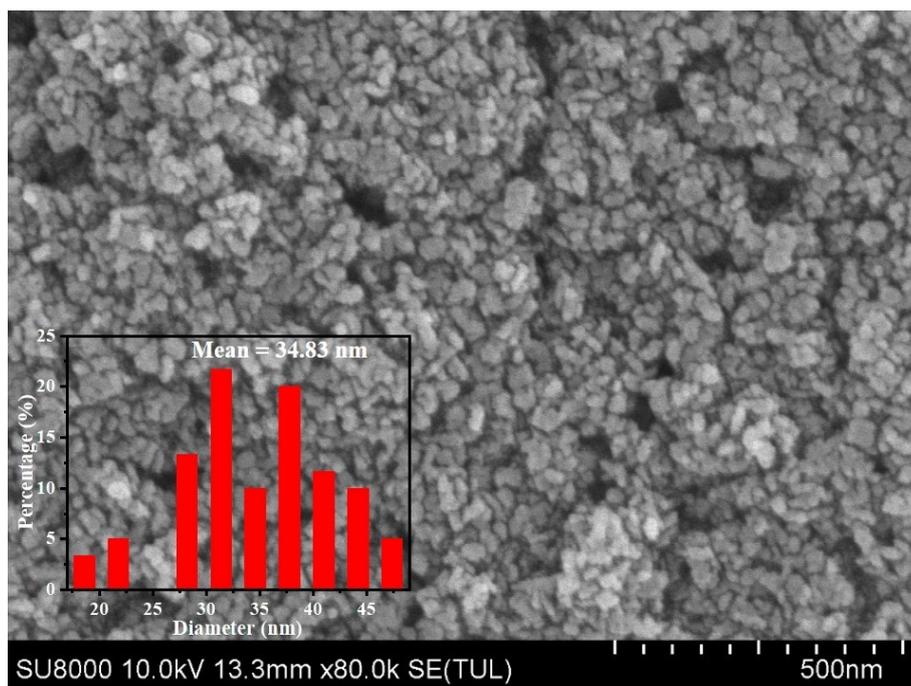


Fig. S5. The SEM image of VOOH nanoparticle prepared at 200 °C, 12 h. ((inset: histogram of size distribution images of the VOOH NPs).

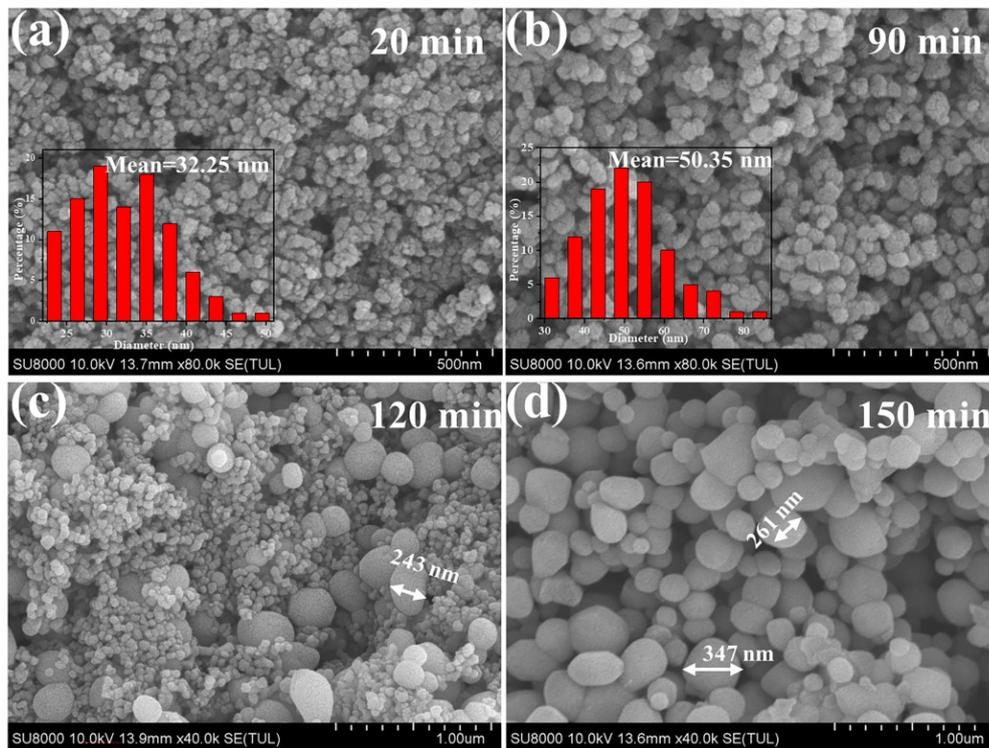


Fig. S6. SEM images of VO₂ with different annealing times: (a) 20 min, (b) 90 min, (c) 120 min and (d) 150 min. (inset: histogram of size distribution of the VO₂ nanoparticles).

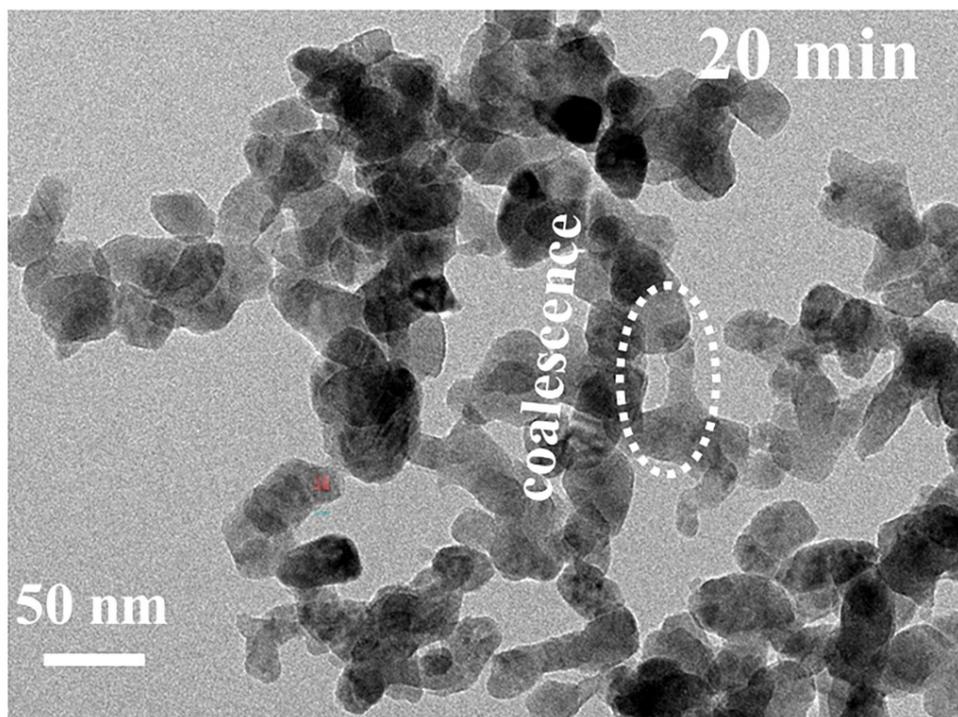


Fig. S7. TEM images of VO₂ with annealing time of 20 min.

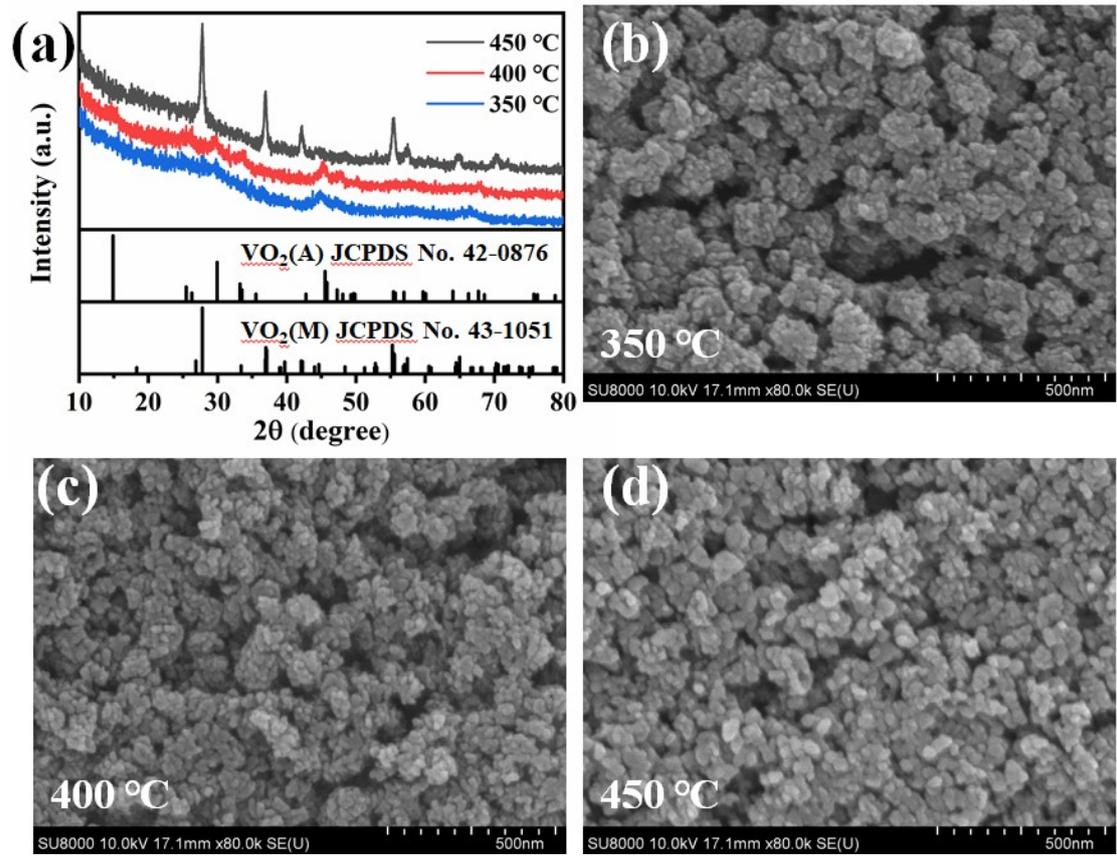


Fig. S8. (a) The XRD patterns of VO₂ with different annealing temperature. The SEM images of annealed samples at temperature of (b) 350 °C, (c) 400 °C and (d) 450 °C.

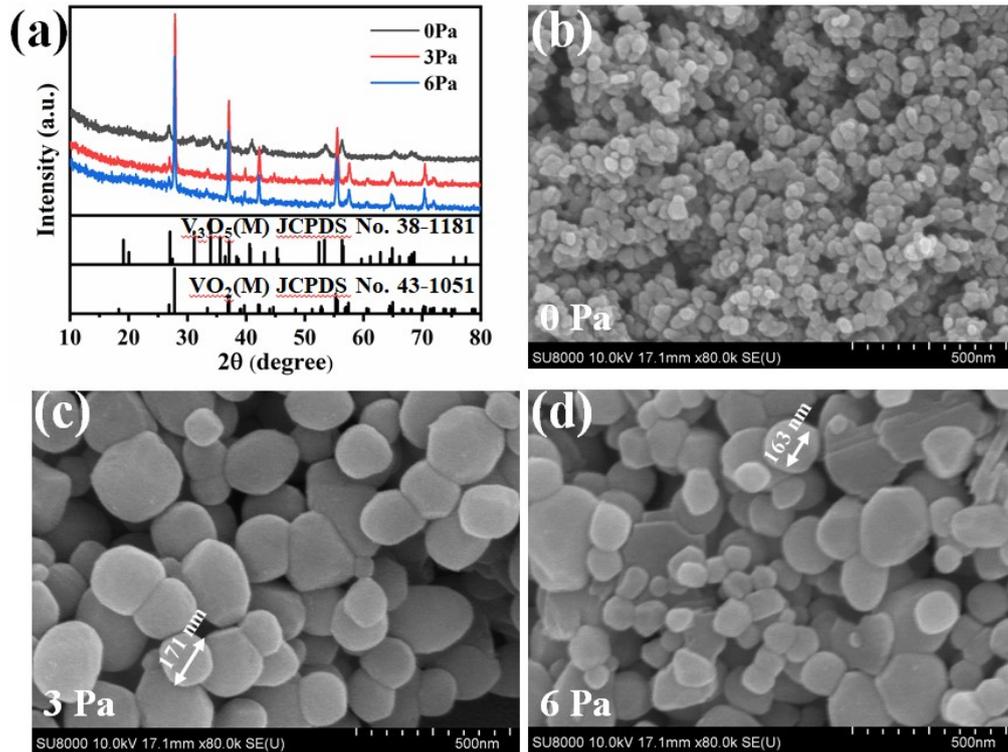


Fig. S9. (a) The XRD patterns of VO_2 with different vacuum degree. The SEM images of annealed samples at vacuum degree of (b) 0 Pa, (c) 3 Pa and (d) 6 Pa.

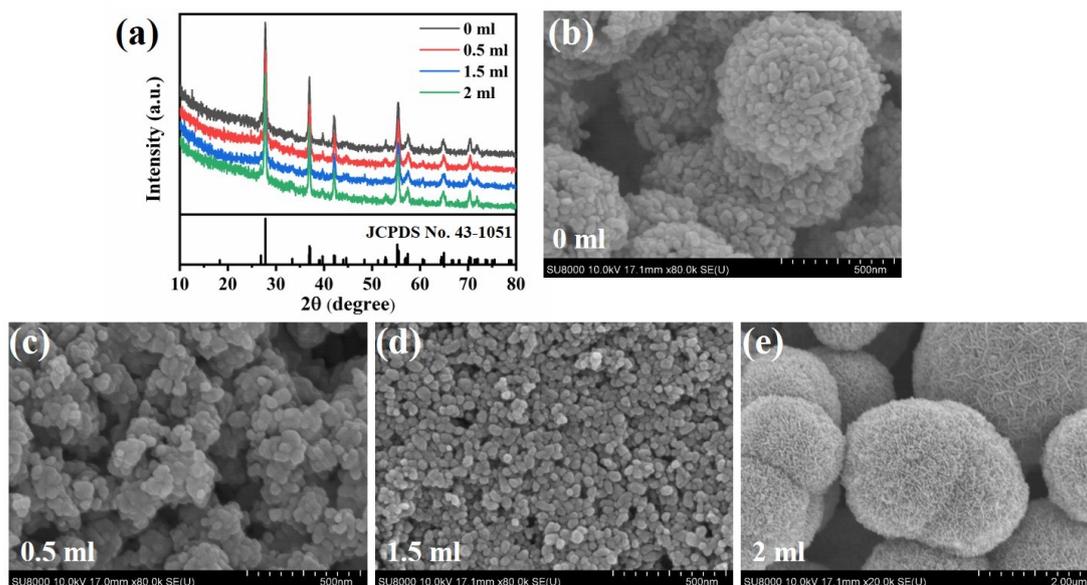


Fig. S10. (a) The XRD patterns of VOOH precursors after annealing treatment. The SEM images of VOOH precursors after annealing treatment with different volume of HCOOH: (b) 0 ml, (c) 0.5 ml, (d) 1.5 ml and (e) 2 ml.

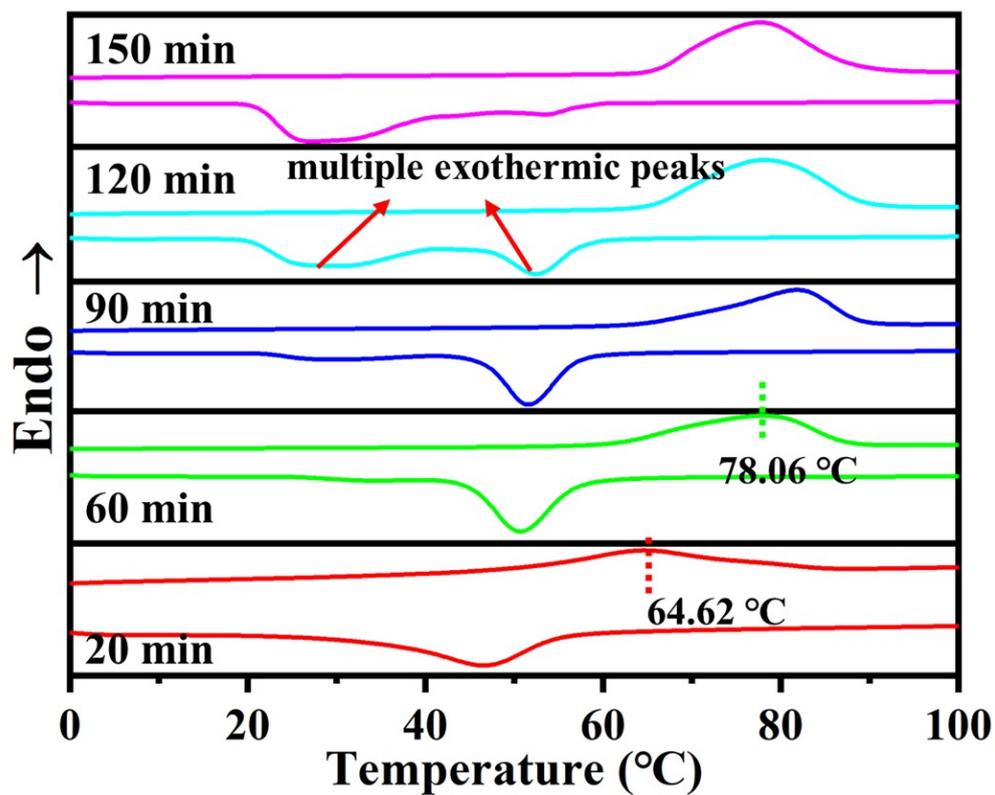


Fig. S11. DSC curves of VO₂ with different annealing times.

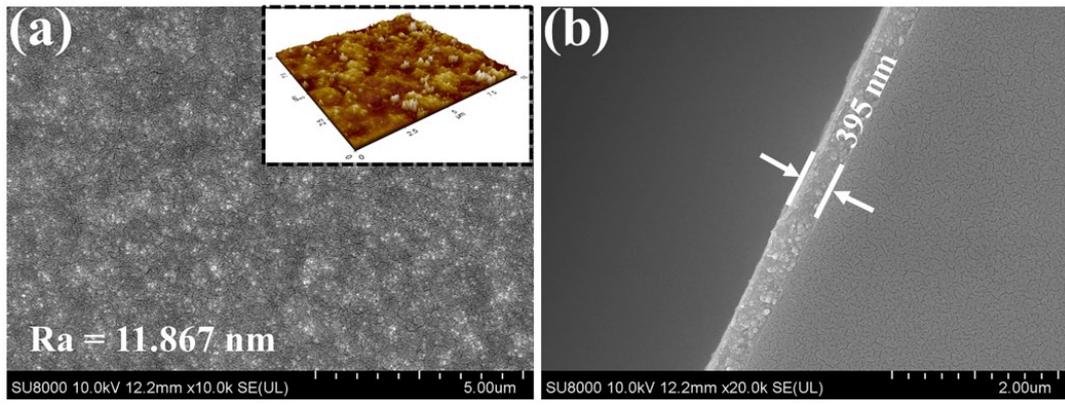


Fig. S12. (a) SEM, AFM images and (b) cross section image of VO₂/PVP-60 min film with spinning coating time of 3

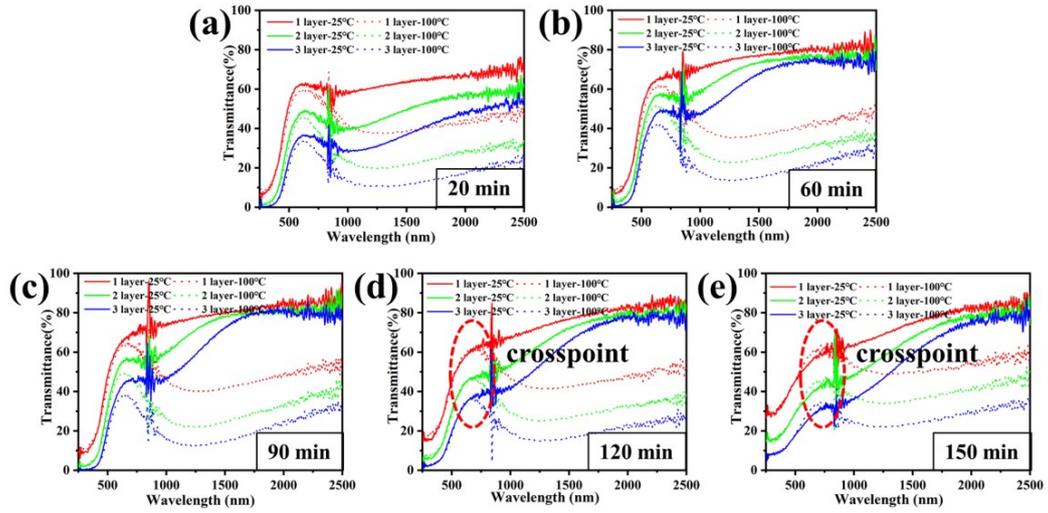


Fig. S13. The transmittance spectra of VO₂/PVP films with different spin coating times: (a) 20 min, (b) 60 min, (c) 90 min, (d) 120 min and (e) 150 min.

Annealing time	Spin coating times	T_{lum} (%)			T_{sol} (%)		
		20 °C	100 °C	T_{lum} (%)	20 °C	100 °C	ΔT_{sol} (%)
20 min	1	59.10	55.65	57.38	54.97	45.97	9.00
	2	43.50	40.56	42.03	40.05	29.62	10.43
	3	32.04	29.07	30.55	29.10	19.67	9.43
60 min	1	58.29	54.61	56.45	60.23	45.27	14.95
	2	48.48	44.41	46.45	50.85	33.70	17.15
	3	38.80	34.73	36.77	42.66	24.58	18.08
90 min	1	57.97	55.49	56.73	62.27	48.22	14.05
	2	45.17	41.40	43.29	50.29	31.84	18.45
	3	33.66	29.75	31.71	40.81	22.00	18.81
120 min	1	52.37	52.71	52.54	58.07	48.20	9.88
	2	38.02	38.24	38.13	45.24	42.43	12.81
	3	27.59	27.77	27.68	36.28	22.61	13.67
150 min	1	50.01	52.15	51.08	56.88	51.85	5.03
	2	33.72	35.73	34.73	42.56	35.88	6.68
	3	22.23	24.47	23.35	31.57	24.47	7.10

Fig. S14. Summarized optical performances of VO₂/PVP films.

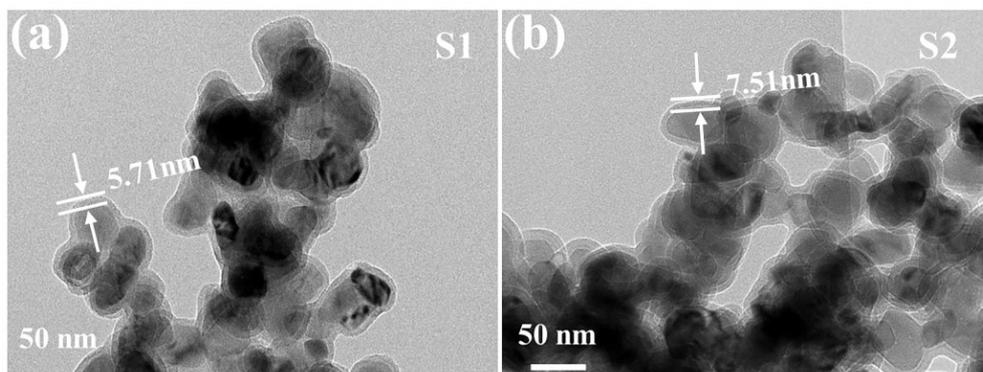


Fig. S15. TEM images of (a) S1, (b) S2.

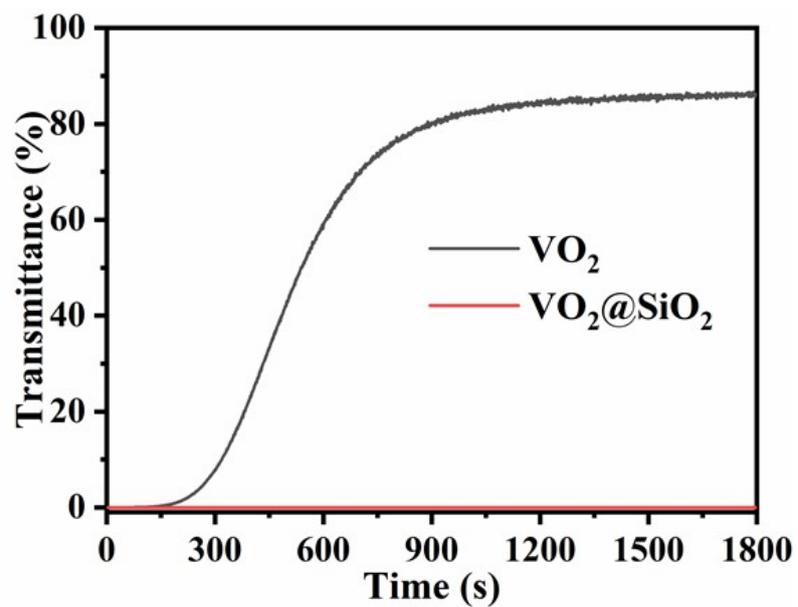


Fig. S16. The time-dependent transmittance at 550 nm of VO₂ and VO₂@SiO₂.

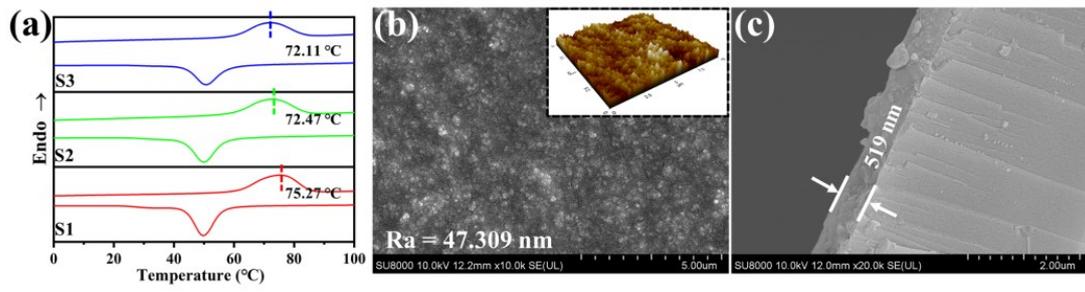


Fig. S17. (a) DSC curves of VO₂@SiO₂ nanoparticles. (b) SEM, AFM images and (c) cross section image of VO₂@SiO₂/PVP film with spin coating time of 3.

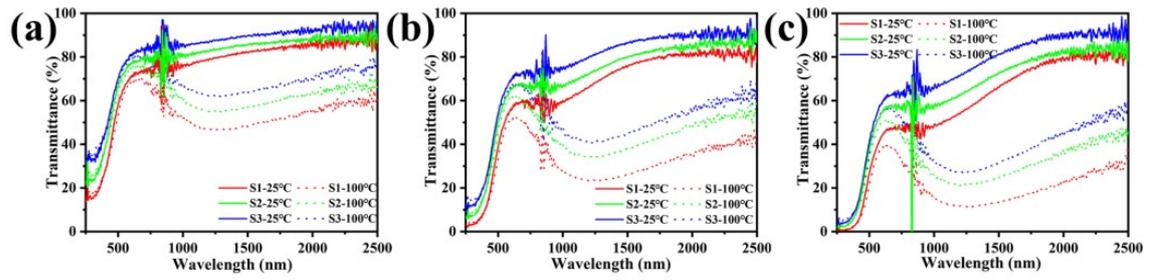


Fig. S18. The transmittance spectra of $\text{VO}_2@SiO_2/PVP$ films with different spin coating times: (a) 1 layer, (b) 2 layer and (c) 3 layer.

Annealing time	Spin coating times	T_{lum} (%)			T_{sol} (%)		
		20 °C	100 °C	T_{lum} (%)	20 °C	100 °C	ΔT_{sol} (%)
S1	1	66.26	63.33	64.79	67.71	55.00	12.71
	2	49.96	45.50	47.73	52.70	34.77	17.92
	3	37.79	32.62	35.20	42.33	22.85	19.48
S2	1	73.36	70.72	72.04	73.63	63.23	10.40
	2	59.79	55.94	57.87	60.80	45.35	15.45
	3	48.81	44.15	46.48	50.53	33.45	17.09
S3	1	77.05	75.19	76.12	78.62	68.65	9.97
	2	63.95	60.64	62.29	66.32	51.40	14.91
	3	52.69	48.69	50.69	57.32	39.02	18.30

Fig. S19. Summarized optical performances of $VO_2@SiO_2/PVP$ films with various spin coating times.

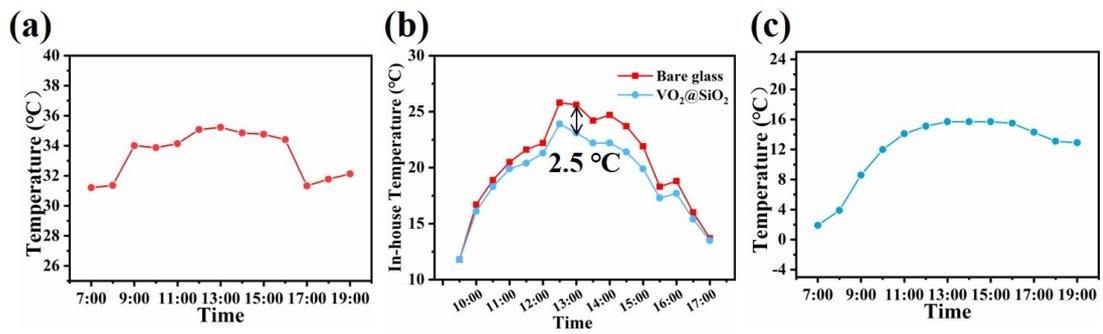


Fig. S20. (a) Temperature change curve in Hefei on 11 July 2023. (b) Temperature vs. time data tested in winter of the model house and (c) temperature change curve in Hefei on 15 November 2023.