## Supplementary Materials for

# Thermally-induced Cyclic Resistance Transition of Transparent and Flameretardant Layered Oxidized MXene Composite Nanocoating for Remote-sync Fire Monitoring

Ye-Jun Wang <sup>1, ‡</sup>, Bi-Fan Guo <sup>1, ‡</sup>, Ling-Yu Lv <sup>1, ‡</sup>, Cheng-Fei Cao <sup>1, \*</sup>, Pei-Yuan Lv <sup>1</sup>, Yang Li <sup>2</sup>,

Guo-Dong Zhang<sup>1</sup>, Jie-Feng Gao<sup>3</sup>, Pingan Song<sup>4</sup>, Kun Cao<sup>5</sup>, Long-Cheng Tang<sup>1,\*</sup>

<sup>1</sup> Key Laboratory of Organosilicon Chemistry and Material Technology, Ministry of Education, Zhejiang Key Laboratory of Organosilicon Material Technology, College of Material, Chemistry and Chemical Engineering, Hangzhou Normal University, Hangzhou, 311121, Zhejiang, China <sup>2</sup> School of Information Science and Technology, Hangzhou Normal University, Hangzhou 311121, China

<sup>3</sup> College of Chemistry and Chemical Engineering, Yangzhou University, Yangzhou, 225002, China
 <sup>4</sup> Centre for Future Materials, University of Southern Queensland, Springfield 4300, Australia
 <sup>5</sup> State Key Laboratory of Chemical Engineering, College of Chemical and Biological Engineering, Zhejiang University, Hangzhou 310027, China

<sup>+</sup>These authors contributed equally to this work.

\* Correspondence: chengfei.cao@unsw.edu.au (C.F. C.), lctang@hznu.edu.cn (L.C. T.).

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#### **Experimental Section**

#### Materials

Ti<sub>3</sub>AlC<sub>2</sub> precursor was purchased from Jilin Technology Co., Ltd. Kevlar fibers was purchased from DuPont USA Co., Ltd (with a diameter of 0.42 mm). Lithium fluoride salt (LiF,  $\geq$ 98 wt%), dimethyl sulfoxide (DMSO,  $\geq$ 99.5 wt%), potassium hydroxide (KOH,  $\geq$ 98 wt%), boric acid (H<sub>3</sub>BO<sub>3</sub>,  $\geq$ 98 wt%), hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>,  $\geq$ 30 wt%) hydrochloric acid (HCl, 35 wt%) and other reagents were bought from Sinopharm Chemical Reagent Co., Ltd. (China). All the chemicals were used without further purification.

#### Preparation of oxidized MXene (OM) sheets

Ti<sub>3</sub>C<sub>2</sub>T<sub>x</sub> MXene sheets were synthesized by the procedure described in previous works. LiF (2 g) and 9 M HCl solution (50 mL) were mixed by stirring at room temperature, followed by the controlled addition of 2 g of MAX. After etching at 40 °C for 48 h, the obtained sample was washed using distilled water for several times until the pH is around 6. The above suspension was further sonicated for 1 h to obtain exfoliated  $Ti_3C_2T_x$  dispersion. To prepare oxidized MXene (OM), hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>) was used as the oxidizing agent. Specifically, 0.2 mL of H<sub>2</sub>O<sub>2</sub> (98 wt%) was added to a 10 mL MXene suspension (5 mg/mL) at room temperature and allowed to react for 20 min. The mixture was then heated at 80 °C to decompose the residual H<sub>2</sub>O<sub>2</sub>. The product obtained under these conditions was labeled as OM<sub>20</sub>. For comparison, additional oxidized MXene samples, designated as OM<sub>10</sub>, OM<sub>30</sub>, OM<sub>60</sub>, and OM<sub>120</sub>, were prepared by varying the oxidation time with H<sub>2</sub>O<sub>2</sub> to 10, 30, 60, and 120 min, respectively.

#### Preparation of aramid nanofibers (ANFs) suspension

In this work, a stable ANFs suspension (Fig. S6b) was prepared using a donor-assisted deprotonation method. A mixture of 1.0 g of KOH, 1 g of aramid fiber, 20 mL of deionized water, and 480 mL of DMSO was stirred magnetically at 1200 rpm for 4 h at room temperature. This process produced a dark red ANFs dispersion with a concentration of 2 mg/mL. The resulting ANFs solution demonstrated excellent stability, with no aggregation or flocculation observed, attributing to electrostatic repulsion. 25 mL ANFs suspension was mixed with 125 mL deionized water, stir it under a magnetic force at 1200 RPM for 2 hours. After filtration, an ANFs pulp with a solid content of 50 mg is obtained (Fig. S6c). The diameter of the prepared ANFs fibers is approximately 150 nm, and the length is 5 µm (Fig. S6d).

#### **Preparation of OMAB**

The ANFs pulp with a solid content of 50 mg, BA (10 mg) and deionized water (20 mL) were added to the  $OM_{20}$  solution (10 g, 5 mg/g) and stirred for 2 h. The resulting uniform suspension was then transferred to a mold and placed in an oven at 50 °C for 4 h. After this low-temperature drying process, OMAB paper were fabricated, where OM represents oxidized MXene, A and B refers to ANFs and BA, respectively. Additionally, MAB papers were also prepared, where M stands for MXene.

#### Wireless operation of visualization system

A visualization system was used for real-time detection of the resistance in the early warning material connected to the wire. The system included the following: (i) commercial resistance detectors for real-time data collection. (ii) Commercial wireless transmitting devices that upload the collected data to web page, the instrument type can be found in the attachment. And (iii) homemade

visual web page designed to process the resistance information further and convert it into visual signals for remote fire monitoring.

#### **Materials Characterizations**

The morphology and microstructure of MXene and various samples were analyzed using scanning electron microscopy (SEM) with an energy-dispersive spectrometer (EDS) on a Sigma-500, ZEISS instrument. Transmission electron microscopy (TEM) images were obtained using a Talos F200X G2 instrument with an accelerating voltage of 200 kV, the burned sample was coated with epoxy resin and then sliced for testing. The chemical compositions and structure of the materials were analyzed using a Nicolet 7000 Fourier-transform infrared (FT-IR) spectrometer and a VG Scientific ESCALab 220I-XL X-ray photoelectron spectrometer (XPS), respectively. X-ray diffraction (XRD) analysis was performed using a Rigaku D/Max 2550 V X-ray diffractor with a 20 range of 5° to 80°. The oxidation behavior of MXene and MXene composites was analyzed via Raman spectroscopy using a spectrometer from Bruker Instruments, Germany, and a universal electricity meter. The thermal performance of the samples was characterized using thermogravimetric analysis (TA Instruments Q500, America) under an air atmosphere with a heating rate of 10 °C/min from room temperature to 700 °C. The thermal infrared images of samples during the burning process were recorded using an infrared camera (FLUCK TI450PRO) with a temperature range from -20 to 900 °C. Electrical resistance of various samples ( $\sim 10 \times 20$  mm) during the burning process was recorded by a multimeter (ESCORT 3146A), and two flexible copper mesh as electrodes were carefully affixed with each end of the above samples. Wireless transmission devices are commercially available.



**Fig. S1** Digital photos of MXene and its derivatives solutions, showing the color transformation of MXene with different oxidation times.



Fig. S2 SEM images of MXene,  $OM_{10}$ ,  $OM_{20}$ ,  $OM_{30}$ ,  $OM_{60}$  and  $OM_{120}$ .



Fig. S3 The variation of solution resistance as a function of oxidation time.



**Fig. S4** (a) FTIR spectra of  $OM_{10}$ ,  $OM_{20}$ ,  $OM_{30}$ ,  $OM_{60}$ ,  $OM_{120}$  and (b) XPS C 1s spectra of  $OM_{120}$  powder. (c) XPS O 1s spectra of  $OM_{20}$  powder.

FT-IR spectra showed peaks at 3437, 1636, 620, 1097 and 1399 cm<sup>-1</sup>, corresponding to absorbed  $H_2O$ , -OH, Ti-O, -C(F)-F and -OH, respectively. The samples of  $OM_{60}$  and  $OM_{120}$  showed the deficiency at 1097 cm<sup>-1</sup>, indicating that  $OM_{60}$  and  $OM_{120}$  were completely oxidized to TiO<sub>2</sub>. And the Ti-C bond in  $OM_{120}$  disappeared in XPS C 1s spectra.

In the O1s spectrum of  $OM_{20}$  (Fig. S4c), it can be clearly observed that there are some original structures of MXene, such as C-Ti-Ox and C-Ti-OH. Combined with the Ti 2p spectrum (Fig. 2k).



Fig. S5 TEM images for (a) MXene, (b) OM<sub>20</sub> and (c) OM<sub>120</sub>.

The TEM images of MXene,  $OM_{20}$ ,  $OM_{120}$  at low magnification reveals presence of the complete nanosheets, nanosheets with holes and particle aggregates lacking complete nanosheets. HRTEM analysis further confirms that the parallel fringe spacing values of  $OM_{120}$  are 0.128 nm, 0.234 nm, and 0.168 nm, corresponding to the crystal planes of anatase (107), anatase (112), and rutile (211) TiO<sub>2</sub> nanoparticles respectively. In comparison to the control group MXene, both  $OM_{20}$  and  $OM_{120}$  exhibit a distinct absence of hexagonal symmetry in their SAED patterns.



**Fig. S6** (a) Digital photos of  $OM_{20}$  powder. (b) ANFs suspension and (c) ANFs after washing and filtering. (d) The SEM image of ANFs fiber (e) UV–vis transmission spectra of OMAB and MAB papers. (f) Transparency of ANFs, OMA and OMAB papers before and after being immersed in water for 7 days. (g) Tensile stress–strain curves of ANFs, OMA and OMAB papers.

The OMAB paper has a transparency of 43.3% and MAB paper is black, and the OMAB paper can maintain transparency<sup>1</sup> after 7 days of immersion in water.



Fig. S7 Surface and cross-sectional SEM images of (a) ANFs, (b) OMA and (c) OMAB papers.



**Fig. S8** (a) Digital photos of ANFs, OMA and OMAB papers before and after being immersed in water for three months, and (b) immersed in DMSO/KOH solution for 36 h. (c) Mechanism diagram of partial dissolution of OMAB in DMSO/KOH solution. (d) the OMAB paper being immersed in various solution environments for two months, showing good structural stability.



**Fig. S9** (a) XPS results of ANFs, OMA and OMAB papers. (b) XPS N 1s spectra of ANFs and OMAB papers. (c) Ti 2p spectra of OMAB network. (d) XPS B 1s spectra of OMAB paper.



Fig. S10 The homemade setup for assessing combustion behaviors of samples.



Fig. S11 Profile photos of PU foam (a) and (b) OMAB@PU after combustion.



Fig. S12 The cross-sectional SEM image of OMAB@W.



Fig. S13 UL-94 tests of (a) wood, (b) ANFs@W, (c) OMA@W and (d) OMAB@W.



**Fig. S14** (a) The self-extinguishing time of wood, ANFs@W, OMA@W and OMAB@W in the UL-94 test. (b) LOI values of wood, ANFs@W, OMA@W and OMAB@W.



Fig. S15 The combustion behaviors of wood ANFs@W, OMA@W and OMAB@W samples were conducted with an oxygen concentration of 30%.



Fig. S16 SEM images of TiO<sub>2</sub> coating after combustion for (a) 30 s, (b) 60 s and (c) 120 s, respectively.



Fig. S17 XRD of OMAB and OMAB after combustion for 30 s, 60 s and 120 s, respectively.

As the combustion time extends, anatase  $TiO_2$  disappears while the quantity of rutile  $TiO_2$  increases. This phenomenon may be due to the conversion of the two crystal types under the combustion of the flame and oxidation of Ti element in  $OM_{20}$ .



**Fig. S18** (a) Cross sectional SEM image of OMAB after 120 s combustion and (b) the corresponding EDS mapping image for B, Ti and N element.



**Fig. S19** Cross-sectional SEM images of OMAB after combustion for (a) 30 s, (b) 60 s and (c) 120 s, respectively.



Fig. S20 (a) TGA results and corresponding (b) DTG curves of ANFs, OMA and OMAB papers.



Fig. S21 XPS B 1s spectra of OMAB paper after combustion for 120 s.



**Fig. S22** Digital photos of flame detection processes of (a) OMAB paper and (b) MAB paper. (c) Schematic illustration of cyclic fire detecting sensor device based on the OMAB paper.



**Fig. S23** (a) The OMAB sensor is fixed to the foam. (b) The blue areas are coated with OMAB on the high-rise burning model. (c) Schematic diagram of wireless transmission of OMAB sensor connected to foam wall and wireless transmission hardware.



**Fig. S24** (a) The monitoring center 150 m away and (b) the homeowner 850 km away in Xiamen synchronously view the fire process.

Samples	<b>t</b> <sub>1</sub> ( <b>s</b> )	<b>t</b> <sub>2</sub> ( <b>s</b> )	$t_{1}+t_{2}(s)$	t <sub>2</sub> +t <sub>3</sub> (s)	Drip or not	UL-94
wood	101				no	NR
ANFs@W	82				no	NR
OMA@W	8	39	47	45	no	V-2
OMAB@W	1	6	7	8	no	V-0

Table S1 UL-94 tests result of wood, ANFs@W, OMA@W and OMAB@W

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Samples	Response time (s)	Transparency	Response temperature (°C)	Recyclable or not	Ref.
PMS/AA coating	~20	×	273	×	2
GO/silicone coating	~3	×	flame	×	3
GO/MXene/CF paper	~2	×	250	×	4
GO/CNTs/CS/WP P paper	~6	×	400	×	5
SA/PEG/Ag NW textile	~3	×	flame	$\checkmark$	6
PEG/MMT/MW CNTs paper	~11	×	flame	×	7
PEPN/BN film	~6	×	200	×	8
PDMS/H <sub>3</sub> BO <sub>3</sub> /BP O/APP/CNTs composite	~5	×	flame	×	9
G/NC film	~4.4	×	232	×	10
OM/ANFs/BA coating	~1.5	$\checkmark$	flame		This work

Table S2 A comparison of comprehensive properties with different fire warning materials.

Notes: PMS: phthalonitrile; AA: acrylic latex; GO: graphene oxide; CF: cellulose; CNTs: carbon nanotubes; CS: chitosan; WPP: wood pulp paper; SA: sodium alginate; PEG: ethylene glycol; Ag NW: silver nitrate; MMT: montmorillonite; MWCNTs: multi-walled carbon nanotubes; PEPN: imide ring via; BN; dimethyl 5-(1,3-dioxoisoindolin-2-yl)isophthalate; PDMS: dimethyl siloxane; BPO: benzoyl peroxide; APP: ammonium polyphosphate; CNTs: carbon nanotubes; G: graphene; NC: nitrocellulose; OM: oxide MXene sheets; ANFs: kevlar fibers; BA: boric acid.

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