# **Supplementary Materials**

# Nanoclay-based 3D aerogel framework for flexible flame retardants

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Movies S1 to S5



Fig. S1 SEM images of (A) R-RT, (B) R-300, (C) R-450 and (D) R-900.



Fig. S2 (A) Aerogel without PA; B Aerogels without synthesized PA in situ



Fig. S3 XRD patterns of cellulose nanocrystal, rectorite and CR.



**Fig. S4** (A) Photographs of CR/PA/PVA aerogels with a diameter of 14 cm. (B-D) CR/PA/PVA-RT aerogels can support pressure of 10 kN at 80% strain and quickly recover to its original height. (E-H) Schematics of compression and elasticity mechanism of CR/PA/PVA aerogel.



**Fig. S5** FTIR spectra of rectorite nanoclay. (R100-R1100 refer to rectorite calcined at 100-1100 °C, respectively. Calcination was performed in a laboratory programmable muffle furnace at a heating rate of 10 °C/min. After being kept at the target temperature for 2 h, it was naturally cooled to RT).



Fig. S6 Value of compressive strength obtained based on Fig. 2h at 80% strain.



Fig. S7 FTIR spectra of the CR/PA/PVA aerogels.



Fig. S8 (A) TGA weight loss and (B) DSC curves of CR/PA/PVA aerogels and control aerogel.



**Fig. S9** Photographs of residues after cone calorimeter tests for (A) C/PA/PVA, (B) CR/PA/PVA-RT, and (C) CR/PA/PVA-900.



**Fig. S10** Photographs of alcohol lamp combustion experiments of (A) C/PA/PVA, (B) CR/PA/PVA-RT, and (C) CR/PA/PVA-900. Magnification corresponding to inset of Fig. 5.



**Fig. S11** FTIR spectra of cellulose nanocrystals, rectorite, PA, PVA, CR/PA/PVA-RT aerogel, and CR/PA/PVA-RT aerogels after cone calorimeter tests.

Samula	Ignition time	gnition time Peak heat-release rate Total heat release		Total smoke release	
(s)		$(kW/m^2)$	$(MJ/m^2)$	$(m^2/m^2)$	
C/PA/PVA	24	37.1	5.69	522.7	
CR/PA/PVA-RT	249	12.9	1.3	72.6	
CR/PA/PVA-300	273	14.2	1.4	49.4	
CR/PA/PVA-450	No flame	8.3	2.0	77.6	
CR/PA/PVA-900	126	13.6	1.2	338.8	

Table S1 Cone calorimeter data of obtained aerogels

Samula	Density	Ignition time	Peak heat-release	Dafaranaa
Sample	$(g/cm^3)$	(s)	rate (kW/m <sup>2</sup> )	Reference
PVA/MMT	0.093±0.003	4	140.1	(1)
PVA/MMT/AP	0.115		115	(2)
PVA/MMT/AP	$0.097 \pm 0.001$	6	80.4	(3)
PVA/MMT/gelatin	$0.108 \pm 0.006$	3	122	(4)
PVA/SA/MMT/boric acid/CaCl2	0.306±0.012	14±1	82.1	(5)
SA/MMT	$0.096 \pm 0.002$	192	19.3	(6)
SA/MMT	$0.098 \pm 0.001$	215	22.7	(7)
Polyimide/graphene oxide/MMT	$0.090 \pm 0.002$	4	52.5	(8)
Xanthan gum/MMT	$0.056 \pm 0.001$		55.2	(9)
SA/MMT	$0.11 \pm 0$	No flame	20	(10)
CR/PA/PVA-RT	0.105	249	12.9	This work
CR/PA/PVA-450	0.998	No flame	8.3	This work

Table S2 Comparison of mechanical properties and flame retardancy of CR/PA/PVA aerogels with other clay-based composite aerogels

Notes: MMT, montmorillonite; PVA, poly(vinyl alcohol); AP, ammonium polyphosphate; SA, sodium alginate

Sample	Ignition time (s)	Peak heat-release rate (kW/m <sup>2</sup> )	Compression strength (kPa)	Reference
MFR/pectin	27	80.1		(11)
PFR/SiO <sub>2</sub>		19±1	150	(12)
Cellulose composite	9	58.51	1600	(13)
Co <sub>3</sub> O <sub>4</sub> -graphene/epoxy resin		329		(14)
PP/fullerene and carbon nanotubes		400±25		(15)
CR/PA/PVA-RT	249	12.9	178.8	This work
CR/PA/PVA-450	No flame	8.3	42	This work

Table S3 Comparison of mechanical properties and flame retardancy of CR/PA/PVA aerogels with other types of materials

Notes: MFR, melamine-formaldehyde resin; PP, polypropylene; PFR, phenol-formaldehyde resin

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#### Movies S1 to S5

Movie S1. Demonstration of the excellent compression resilience of CR/PA/PVA composite aerogels.

**Movie S2.** Demonstration that the CR/PA/PVA aerogel with a diameter of 14 cm can withstand a pressure of 10 kN and has good resilience.

Movie S3. Demonstration of the burning process of CR/PA/PVA composite aerogel on a cone calorimeter.

Movie S4. Demonstration of the burning process of C/PA/PVA composite aerogel on a cone calorimeter.

**Movie S5.** Demonstration of three kinds of aerogels being combusted on an alcohol lamp, including C/PA/PVA, CR/PA/PVA, and CR/PA/PVA-900.