

## Supporting Information

### Engineering flame-retardancy in water-based adhesives: A structure–performance driven approach for enhanced safety and functionality.

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Sr. No	Content
S1	Additional Experimental Details.
Table S1	FTIR Peak Assignments.
Table S2	XRD Data Table.
Table S3	Thermogravimetric Data Table.
Table S4	Effect of PAH@TS-2 coating add-on on LOI of cotton fabric.
Table S5	LOI data table.
Fig. S1	Weight bearing test.
Fig. S2	Underwater load bearing test.

#### S1. Additional experimental details

Polyvinyl alcohol (PVA), tannic acid (TA), sodium hexametaphosphate (SHMP), and aluminium hydroxide (Al(OH)<sub>3</sub>) were employed without additional purification. Adhesive mixtures were formulated following the procedure detailed in the main manuscript. The adhesive layer was applied uniformly to wooden substrates (100 mm × 25 mm × 3 mm) using a film applicator, and the bonded assemblies were cured at 60 °C for 24 h.

Water contact angle measurements were conducted using a goniometer (Apex ACAMNSC08, India) via the sessile drop method with 2 μL droplets under ambient laboratory conditions. Tensile shear adhesion testing was performed with a universal testing machine (Tinius Olsen H25KT, India) in accordance with ASTM D906-20 at 28 ± 2 °C and 65–70% relative humidity.

Wet adhesion performance was assessed by immersing bonded samples in distilled water at 28 ± 2 °C for 2 h, followed by mechanical testing immediately after removal to simulate adhesion under humid or moisture-exposed service conditions.

Table S1. Key FTIR peak assignments of components and PAH@TS adhesive.

Sample	Wavenumber (cm <sup>-1</sup> )	Assignment
TA	3251	O–H stretching
TA	1709	C=O (ester/carboxyl)
SHMP	3390	O–H (hydrated phosphate)
SHMP	1090	P–O–P
PVA	3281	O–H stretching
PVA	1095	C–O–C
PAH@TS	3302	Hydrogen-bonded O–H
PAH@TS	1094	P–O / P–O–P

Table S2. XRD peak positions of PAH@TS adhesive.

2θ (°)	Assignment
19.9	PVA semi-crystalline (101)
22.6	Cellulosic substrate contribution
31.9	TS structural ordering
34.1	Phosphate-related domain
37.9	ATH transition
46.1	Amorphous crosslinked phase

Table S3. TGA weight loss profile.

Sample	Temperature range (°C)	Weight loss (%)	Interpretation
PVA	65–133	8	Moisture loss
PVA	260–428	83	Polymer backbone degradation
PVA	428–516	~100	Complete decomposition
PAH	70–165	0.5	Bound water
PAH	338–575	68.9	Polymer degradation
P@TS	71–306	10	Dehydration + initial phosphate activity
P@TS	316–800	68	Carbonization

PAH@TS	61–330	7	Moisture + early char formation
PAH@TS	461–800	56.9	Stable char residue

Table S4. Effect of PAH@TS-2 coating add-on on LOI of cotton fabric.

Loading	LOI (%)
15 %	31.2 ± 0.264
20 %	36.8 ± 0.659
25 %	47.31 ± 0.831
30 %	48.2 ± 0.515

Table S5. LOI values of adhesive materials.

Material (25 % loading on cotton fabric)	LOI (%)
PVA	18.5. ±
PAH	20.6. ±
P@TS	36.7 ±
PAH@TS-1	38.7 ±
PAH@TS-2	47.3 ±
PAH@TS-3	48.5 ±

**Fig. S1.** Weight-bearing evaluation of the PAH@TS adhesive. A wood–wood bonded specimen supporting a static **20 kg load** for 10 min without failure, demonstrating robust adhesion performance compared to other formulations.



**Fig. S2.** Underwater load-bearing test of the PAH@TS adhesive. A bonded wooden substrate was fully submerged in distilled water, and a 200 g metallic load was suspended from the joint to evaluate wet adhesion stability. The joint remained intact for an extended duration, indicating robust underwater bonding performance.

