Supplementary Information for

Tailoring chain-packing structure in co-blended aramid composites for high mechanical and insulating performance

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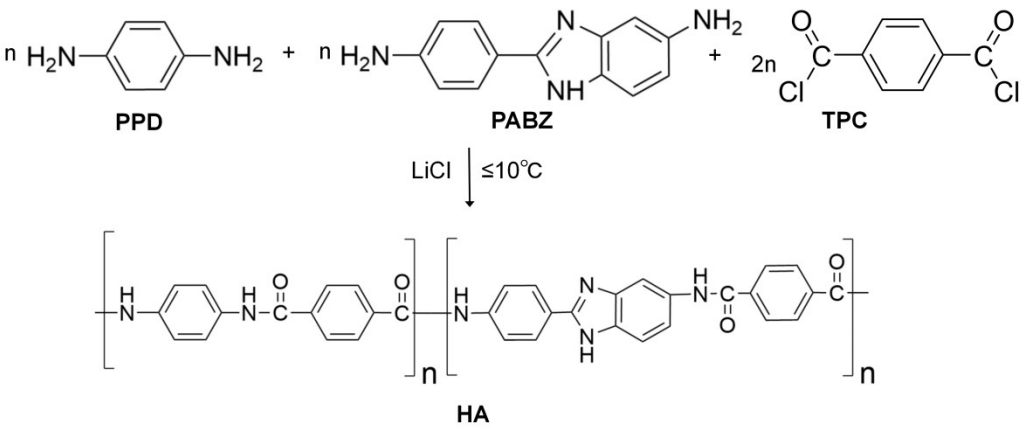

Figure S1. The reaction process among PPD, PABZ, and TPC for the preparation of HA.

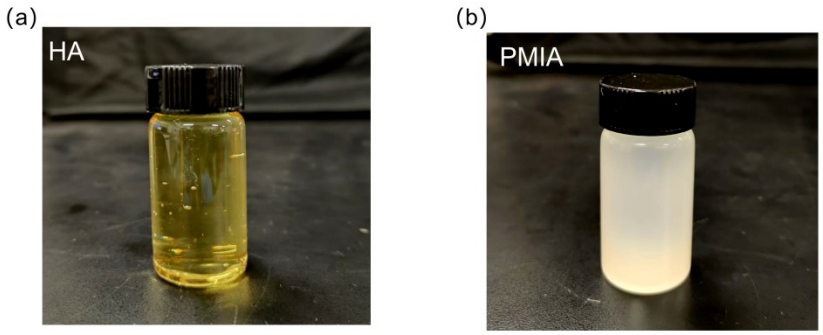

Figure S2. (a) HA in DMAC/LiCl and (b) PMIA in DMAC/LiCl.

Table S1. The dimensions of the paper samples used for each characterization.

Characterizations	Dimensions		
	Length (cm)	Width (cm)	Thickness (mm)
SEM	1.00	1.00	~0.15
FTIR	1.00	1.00	~0.15
XRD	1.00	1.00	~0.15
Breakdown strength	5.00	5.00	~0.15
Volume conductivity	10.00	10.00	~0.15
Charge trap distribution	3.00	3.00	~0.15
Dielectric properties	5.00	5.00	~0.15
Mechanical properties	10.00	1.00	~0.15

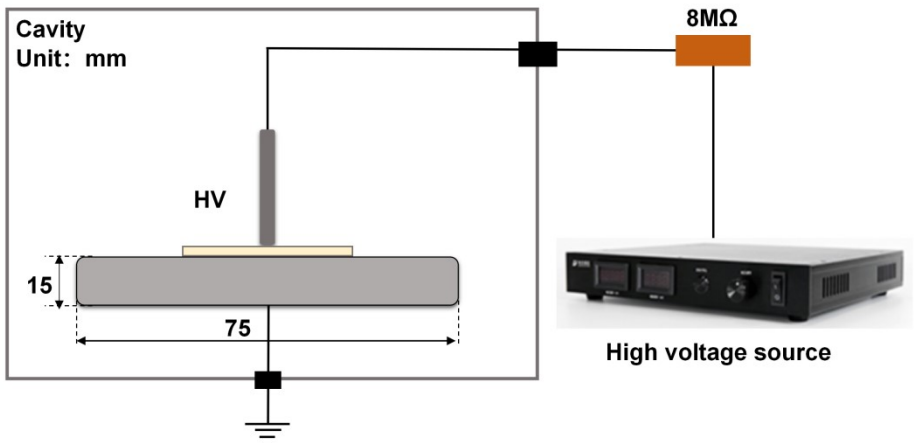

Figure S3. The breakdown testing platform.

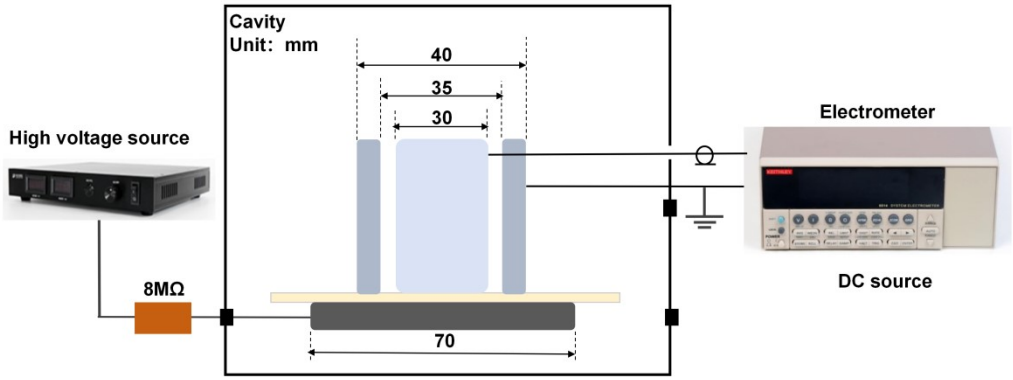

Figure S4. The volume conductivity testing platform.

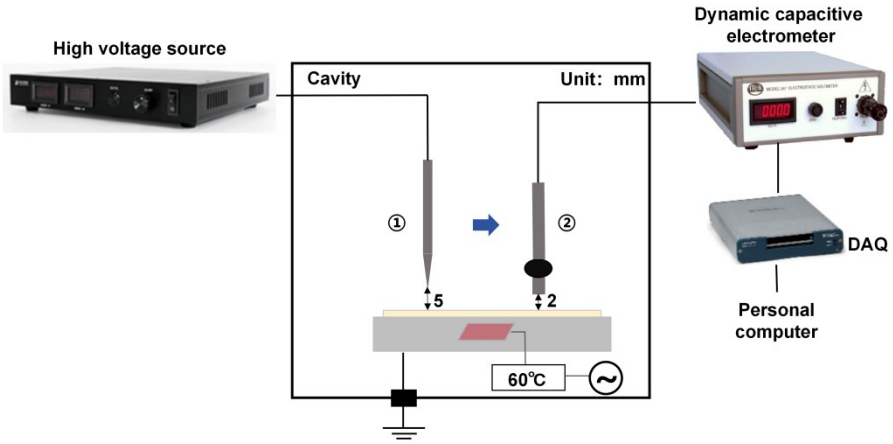

Figure S5. The ISPD testing platform.

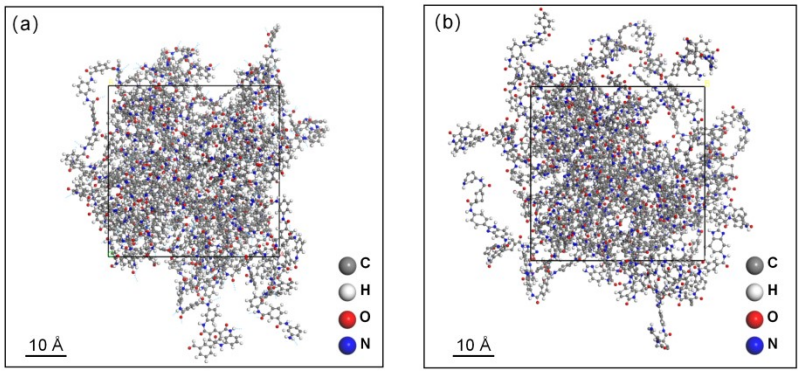

Figure S6. (a) Pure PMIA and (b) HA models.

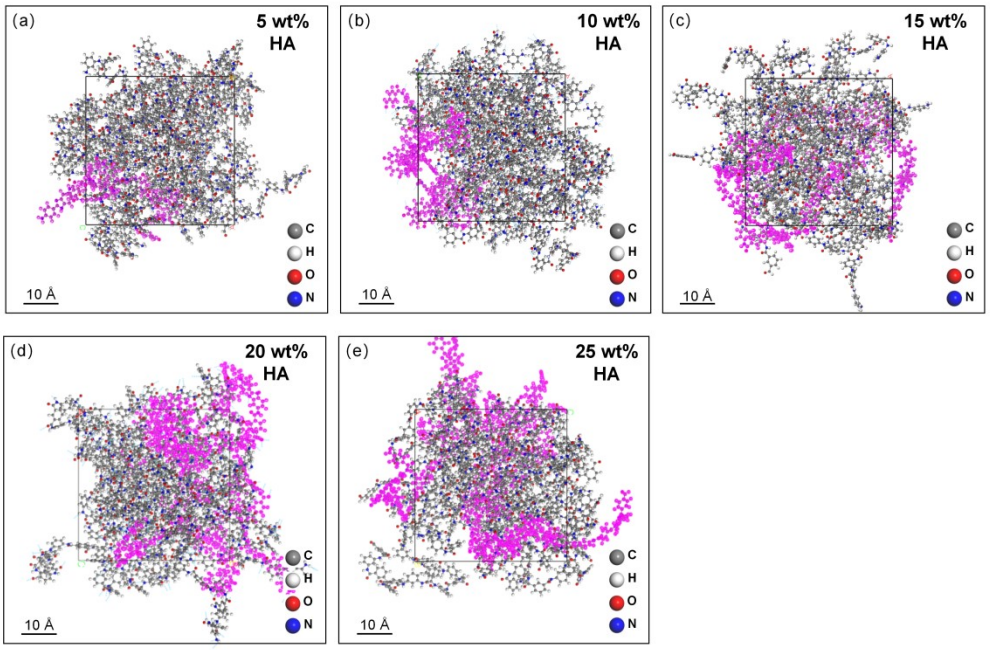

Figure S7. The PMIA/HA co-blending models with the HA content being (a) 5 wt%, (b) 10 wt%, (c) 15 wt%, (d) 20 wt%, and (e) 25 wt%. HA molecular chains are colored in pink.

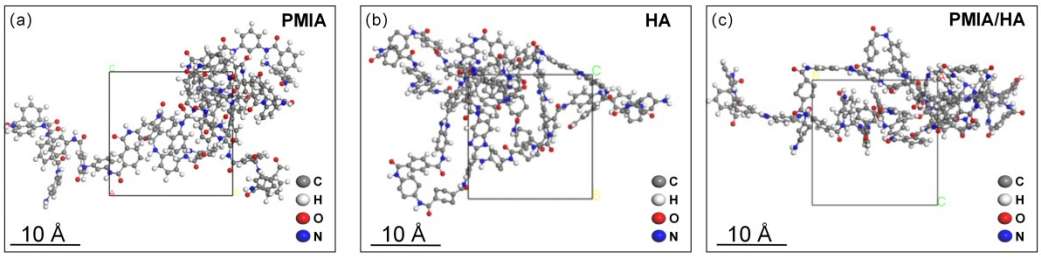

Figure S8. (a) Bi-molecular chain models of PMIA-PMIA, (b) HA-HA, and (c) PMIA-HA.

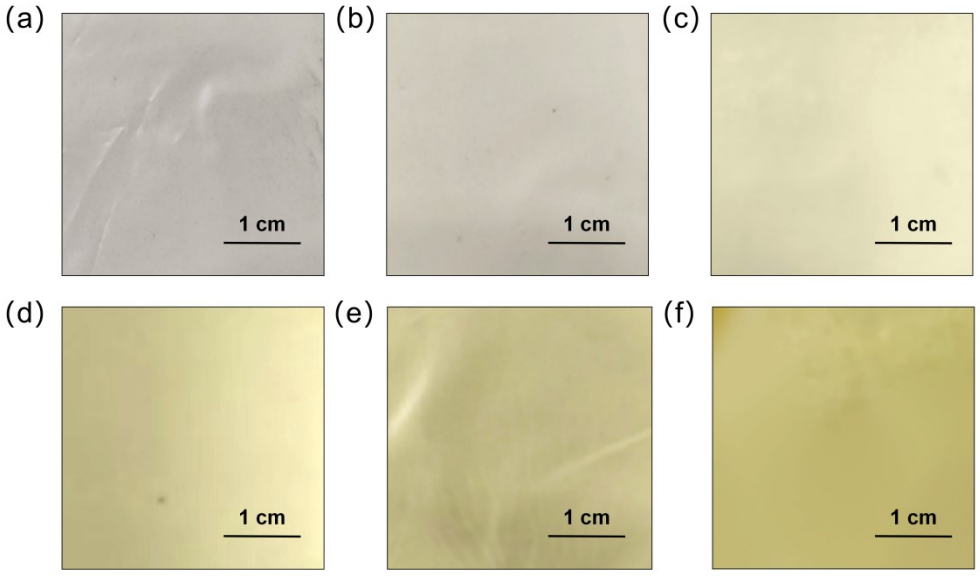

Figure S9. The photographic images of PMIA/HA co-blending papers with the HA content being (a) 0 wt%, (b) 5 wt%, (c) 10 wt%, (d) 15 wt%, (e) 20 wt%, and (f) 25 wt%.

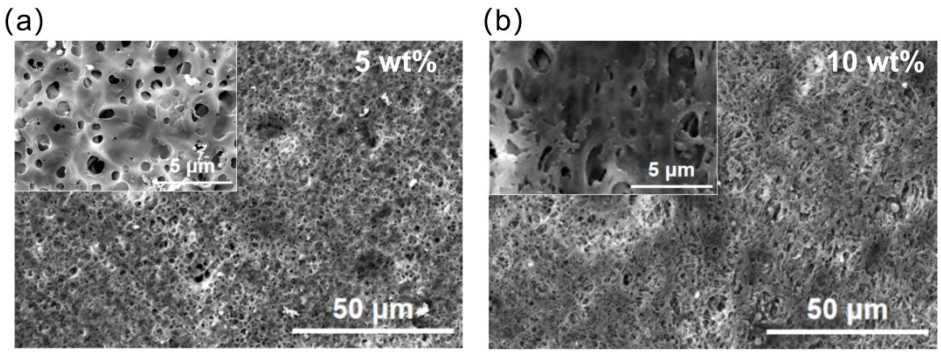

Figure S10. (a) Surface morphology of the 5 wt% PMIA/HA paper and (b) the 10 wt% PMIA/HA paper.

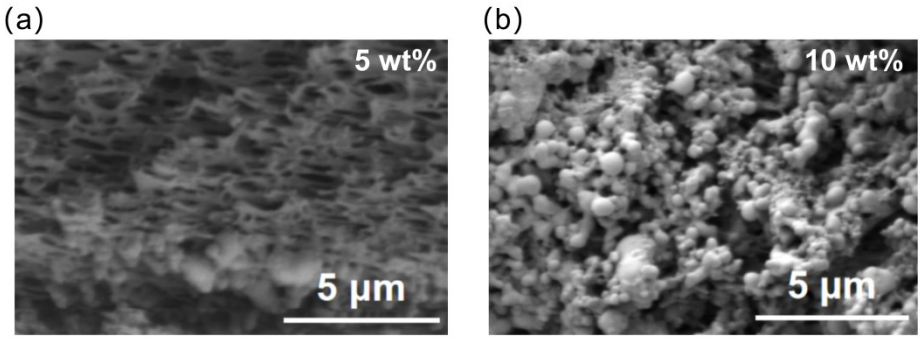

Figure S11. (a) Cross-section SEM images of the 5 wt% PMIA/HA paper and (b) the 10 wt% PMIA/HA paper.

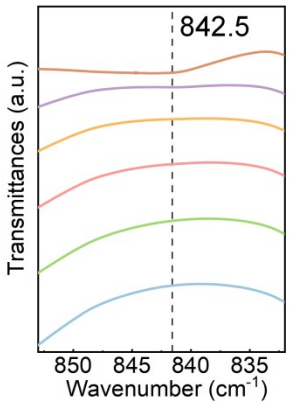

Figure S12. FTIR spectra of various co-blending papers with the HA content being 0 wt%, 5 wt%, 10 wt%, 15 wt%, 20 wt%, and 25 wt%, from the bottom up.

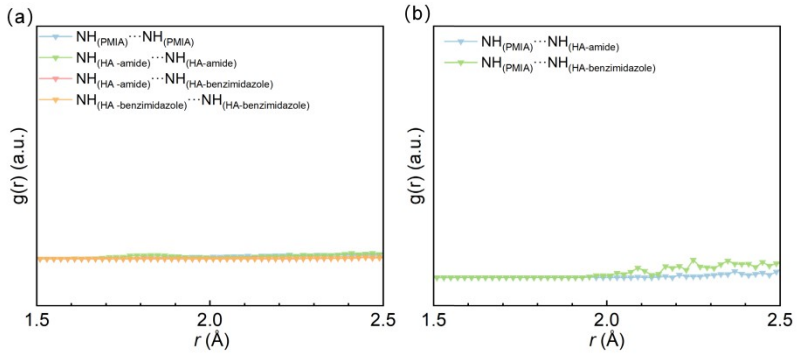

Figure S13. (a) RDF spectra extracted from the pure PMIA and HA model, and (b) extracted from the 20 wt% co-blending model.

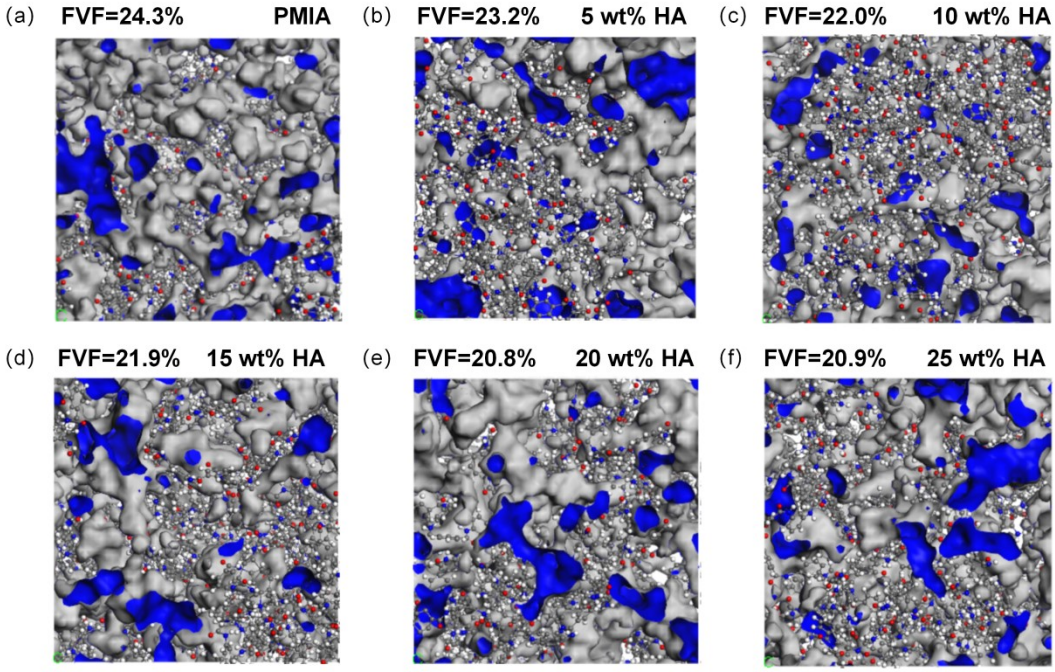

Figure S14. Free volume fraction (FVF) of PMIA/HA co-blending models with the HA content being (a) 0 wt%, (b) 5 wt%, (c) 10 wt%, (d) 15 wt%, (e) 20 wt%, and (f) 25 wt%. The free volume region is indicated by blue color.

Note 1. The method and process of DFT simulations

Individual monomer models of PMIA and HA are constructed. Density Functional Theory (DFT) is employed to evaluate the Lowest Unoccupied Molecular Orbital (LUMO) energy levels and dipole moments of two models. The DFT calculations utilize the B3LYP hybrid function, paired with the 6-31G(d) basis set, which includes polarization functions for improved accuracy in describing molecular structures and interactions. B3LYP, an abbreviation for Becke, 3-parameter, Lee-Yang-Parr, combines Hartree-Fock methods with gradient-corrected functions to enhance the calculation accuracy. The 6-31G(d) basis set is a split-valence set that provides two basis functions per atom to describe valence electrons and includes d-orbital polarization functions. The LUMO levels and dipole moments of the two models are obtained. The optimized monomer models are depicted in Figure S15.

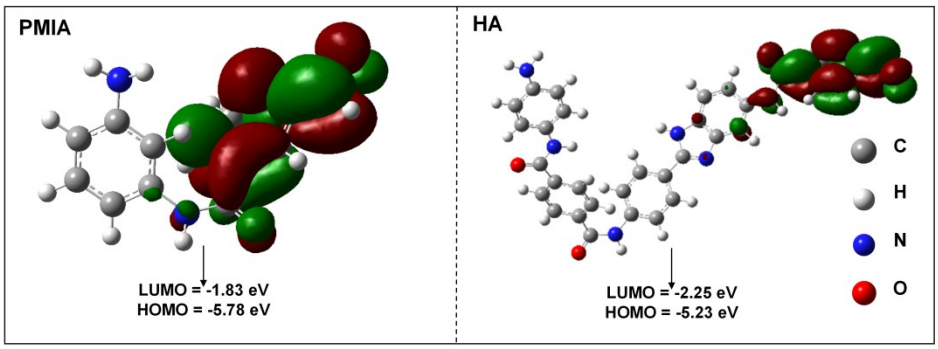

Figure S15. (a) PMIA monomer and (b) HA monomer.

Table S2. The data points for dielectric properties.

HA doping ratio (wt%)		0	5	10	15	20	25
Dielectric constant	Sample set 1	2.809	2.799	2.835	2.921	2.811	2.854
	Sample set 2	2.884	2.987	2.966	2.971	2.981	2.964
	Sample set 3	2.959	2.893	2.902	2.863	2.896	2.909
Dielectric loss	Sample set 1	0.01292	0.01210	0.01189	0.01157	0.01098	0.01122
	Sample set 2	0.01313	0.01249	0.01216	0.01185	0.01139	0.01154
	Sample set 3	0.01271	0.01178	0.01189	0.01124	0.01057	0.01090

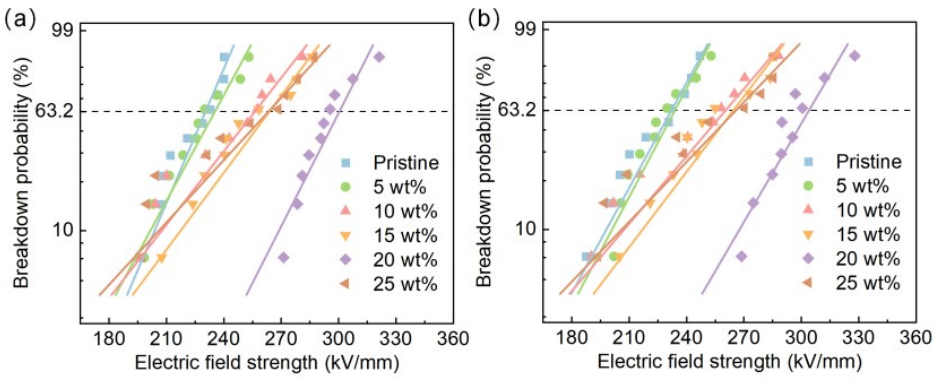

Figure S16. (a) Weibull distribution plots of measured breakdown strengths for sample set 2 and (b) sample set 3.

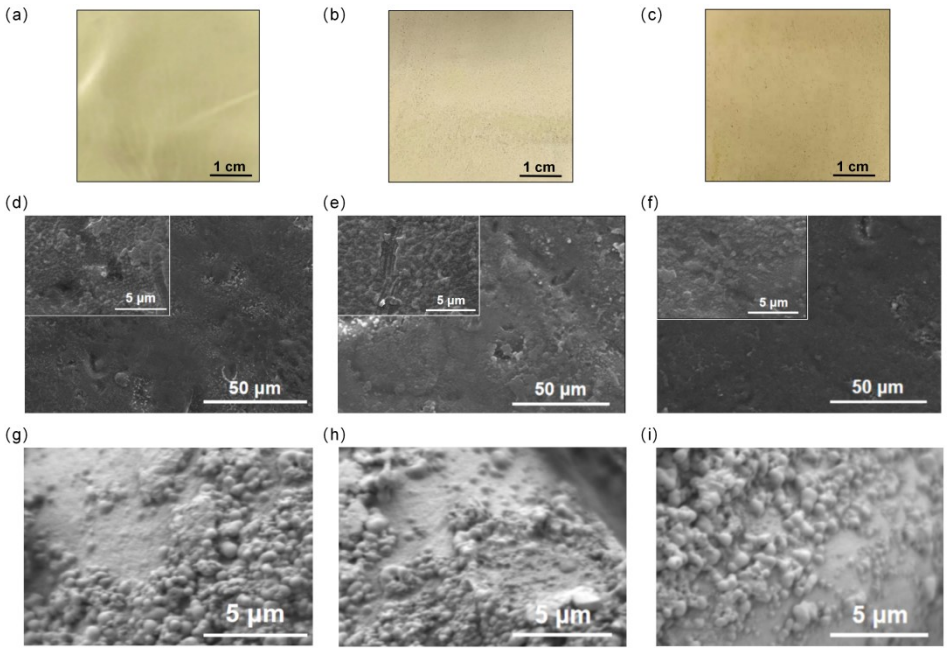

Figure S17. Morphologies of 20 wt% PMIA/HA co-blending papers. (a)-(c) Photographic images, (d)-(e) Surface SEM images, and (g)-(i) cross-sectional SEM images. (a), (d), and (g) from the Main text; (b), (e), and (g) acquired from the sample in Batch 2; (c), (f), and (i) acquired from the sample in Batch 3.

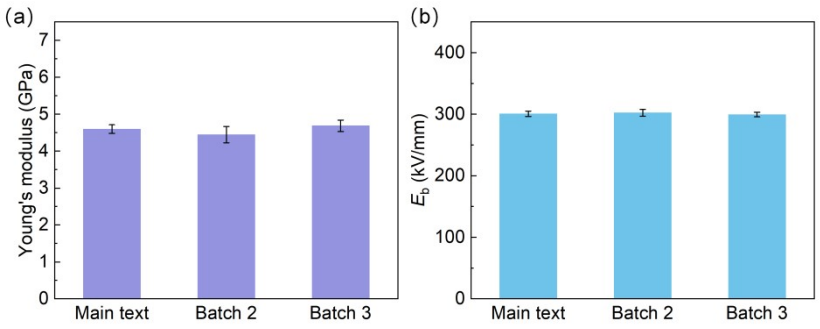

Figure S18. (a) Young's modulus and (b) E_b values of PMIA/HA co-blending paper in the main text and samples from two additional batches.