Electronic Supplementary Material (ESI) for Journal of Materials Chemistry C.

Supporting Information

Novel, Flexible, and Transparent Thin Film Polyimide Aerogels with Enhanced Thermal Insulation and High Service Temperature

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List of Abbreviation

2D	Two-dimensional	PI	Polyimide
3D	Three-dimensional	PINF	PI nanofibers
6FAPB	2,2-bis(3,4-dicarboxyphenyl)hexafluoropropane dianhydride	PMDA	Pyromellitic dianhydride
BAP4	1,4-bis(4-aminophenoxy)butane	PMN	Endic anhydride maleic anhydride copolymer
BAP6	1,6-bis(4-aminophenoxy)hexane	POSS	Polyhedral oligomeric silsesquioxane
BAP10	1,10-bis(4-aminophenoxy)decane	PPDA	P-phenylenediamine
BAPN	1,3-bis(4-aminophenoxy)-2,2-dimethylpropane	PSMA	Styrene-maleic anhydride copolymer
BPDA	Biphenyl-tetracarboxylic acid dianhydride	PTFE	Polytetrafluoroethylene
BTC	1,3,5-benzenetricarbonyl tri-chloride	PU	Polyurethan
BTFB	2,2'-Bis(trifluoromethyl)benzidine	PVDF	Polyvinylidene fluoride
DADD	1,12-dodecyldiamine	TAB	1,3,5-triaminophenoxybenzene
CBDA	1,2,3,4- cyclobutanetetracarboxylic dianhydride	TEA	Triethylamine
DMBZ	3,3′-dimethylbenzidine	TFDB	2,2-bis(trifluoromethyl)-4,4-di-aminobiphenyl
CNF	Cellulose nanofiber	TFMB	2,2·-bis-(trifluoromethyl)-4,4·-diaminobiphenyl
FEP	Poly(tetrafluoroethylene-co-hexafluoropropylene)	TGA	Thermogravimetric analyzer
FPEN	Fluorinated polyarylene ether nitrile	TPU	Thermoplastic polyurethane
FTIR	Fourier-transform infrared spectroscopy	WCA	Water contact angle
MSQ	Poly(methyl silsesquioxane)		
NMP	N-methylpyrrolidinone		
OAPS	Octa(aminophenyl)silsesquioxane		
ODA	4,4′-oxydianiline		
ODPA	4,4'-oxydiphthalic dianhydride		
PAA	Polyamic acid		
PABZ	2-(4-aminophenyl)-5-aminobenzimidazole		
PC	Personal computer		

Table S1. Summary of PI aerogel films reported in literature

Diamine	**Dianhydride**	**Crosslinker**	**Fabrication Method**	**Drying Technique**	**Density (g/cm^3)**	**Surface Area (m^2/g)**	**Thermal conductivity (mW/m.K)**	**Onset of decomposition (℃)**	**Transparent (yes/no)**	**Flexible (yes/no)**	**Application**	**Ref**
PPDA	BPDA	BTC	Mold casting	SCD	0.323-0.424	244.5-380.5	NA	606	Yes	NA	TENG	[1]
ODA	BPDA	TAB	Mold casting	SCD	0.108-0.235	177-383	NA	NA	No	Yes	Filtration	[2]
ODA/DMBZ	BPDA	OAPS	Doctor Blade	SCD	NA	212	NA	NA	No	Yes	TENG	[3]
ODA/BAPx	BPDA	TAB	Mold casting	SCD	0.110-0.299	191-445	NA	496-515	No	Yes	Substrate for lightweight antenna	[4]
DMBZ/BAPN	BPDA	TAB	Mold casting	SCD	0.079-0.304	239-490	NA	NA	No	Yes	Substrate for lightweight antenna	[5]
DMBZ/DADD	BPDA	TAB	Mold casting	SCD	0.076-0.228	47-482	NA	NA	No	Yes	Substrate for lightweight antenna	[6]
BAX	BPDA	OAPS	Doctor Blade	SCD	NA	NA	14.4	NA	NA	Yes	Insulation in aerospace	[7]
ODA/OTD	PMDA	NA	Mold casting	SCD	0.161-0.332	NA	~35-45	NA	Yes	Yes	Solar thermal collector	[8]
ODA/DMBZ/PPDA	BPDA	OAPS	Doctor Blade	SCD	0.108-0.451	254-507	NA	NA	No	Yes	NA	[9]
BAPP	BTDA	TAB	Doctor Blade	SCD	0.26-0.39	NA	NA	NA	No	Yes	Aerospace, pipe wrapping, clothing	[10]
ODA/DMBZ	BPDA	BTC	Mold Casting, Doctor Blade	SCD	0.089-0.119	NA	27.5-35.8	510-556	Yes	Yes	Thermal management in microelectronics	This work

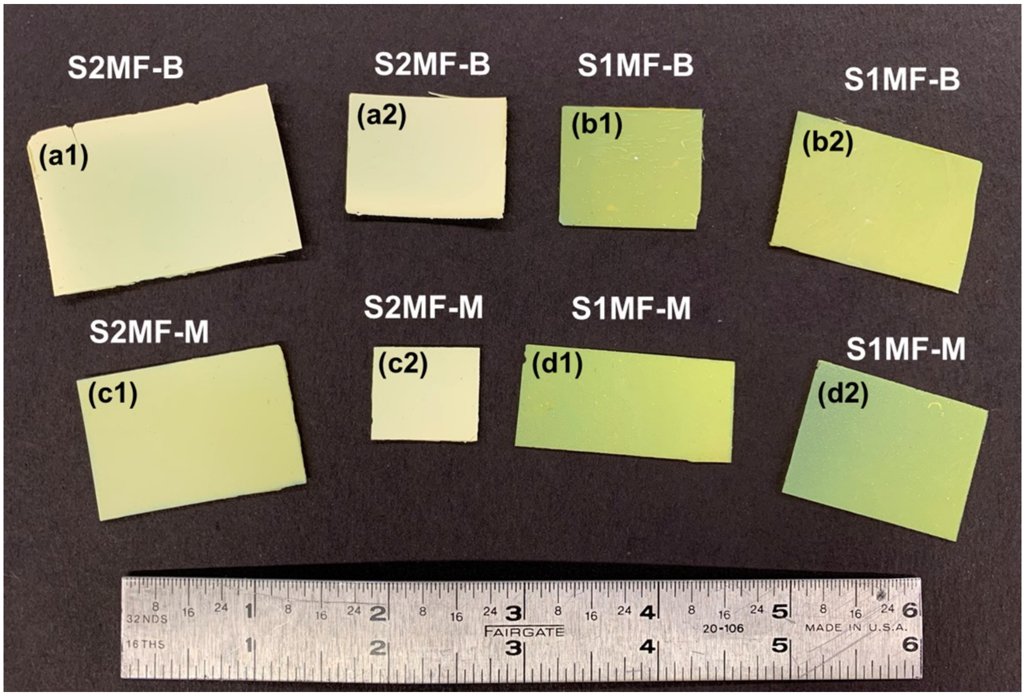
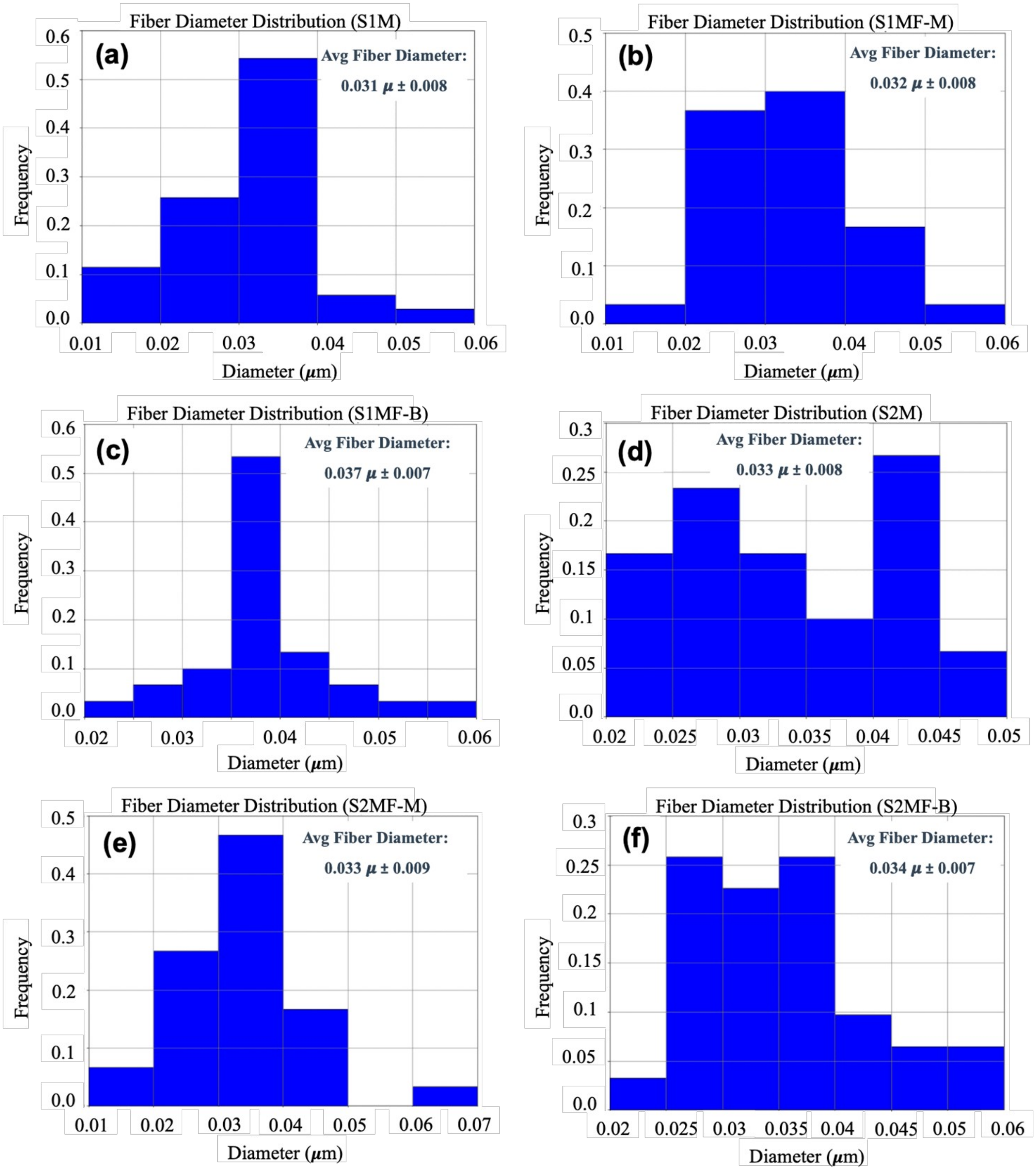


Figure S1. Scalability of PI aerogel thin films: (a1) S2MF-B with 47.9*32.8*0.4 mm^3 dimensions, (a2) S2MF-B with 31.7*27.8*0.4 mm^3 dimensions, (b1) S1MF-B with 28.5*24.3*0.6 mm^3 dimensions, (b2) S1MF-B with 38.7*18.4*0.6 mm^3 dimensions, (c1) S2MF-M with 39.4*24.5*0.9 mm^3 dimensions, (c2) S2MF-M with 21.3*20.3*0.9 mm^3 dimensions, (d1) S1MF-M with 42.2*26.9*0.5 mm^3 dimensions, (d2) S1MF-M with 35.5*27.1*0.5 mm^3 dimensions


Fiber Diameter Distribution (S1M)
(a)
Avg Fiber Diameter:
0.031 μ ± 0.008
Frequency
0.0
0.1
0.2
0.3
0.4
0.5
0.6
0.01
0.02
0.03
0.04
0.05
0.06
Diameter (μm)
Fiber Diameter Distribution (S1MF-M)
(b)
Avg Fiber Diameter:
0.032 μ ± 0.008
Frequency
0.0
0.1
0.2
0.3
0.4
0.5
0.01
0.02
0.03
0.04
0.05
0.06
Diameter (μm)
Fiber Diameter Distribution (S1MF-B)
(c)
Avg Fiber Diameter:
0.037 μ ± 0.007
Frequency
0.0
0.1
0.2
0.3
0.4
0.5
0.6
0.02
0.03
0.04
0.05
0.06
Diameter (μm)
Fiber Diameter Distribution (S2M)
(d)
Avg Fiber Diameter:
0.033 μ ± 0.008
Frequency
0.0
0.05
0.1
0.15
0.2
0.25
0.3
0.02
0.025
0.03
0.035
0.04
0.045
0.05
Diameter (μm)
Fiber Diameter Distribution (S2MF-M)
(e)
Avg Fiber Diameter:
0.033 μ ± 0.009
Frequency
0.0
0.1
0.2
0.3
0.4
0.5
0.01
0.02
0.03
0.04
0.05
0.06
0.07
Diameter (μm)
Fiber Diameter Distribution (S2MF-B)
(f)
Avg Fiber Diameter:
0.034 μ ± 0.007
Frequency
0.0
0.05
0.1
0.15
0.2
0.25
0.3
0.02
0.025
0.03
0.035
0.04
0.045
0.05
0.06
Diameter (μm)

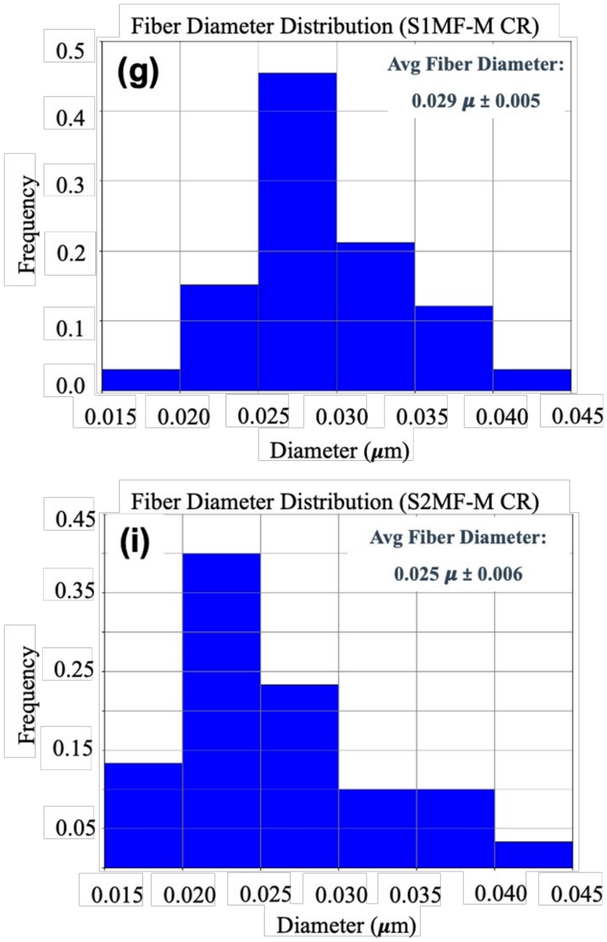
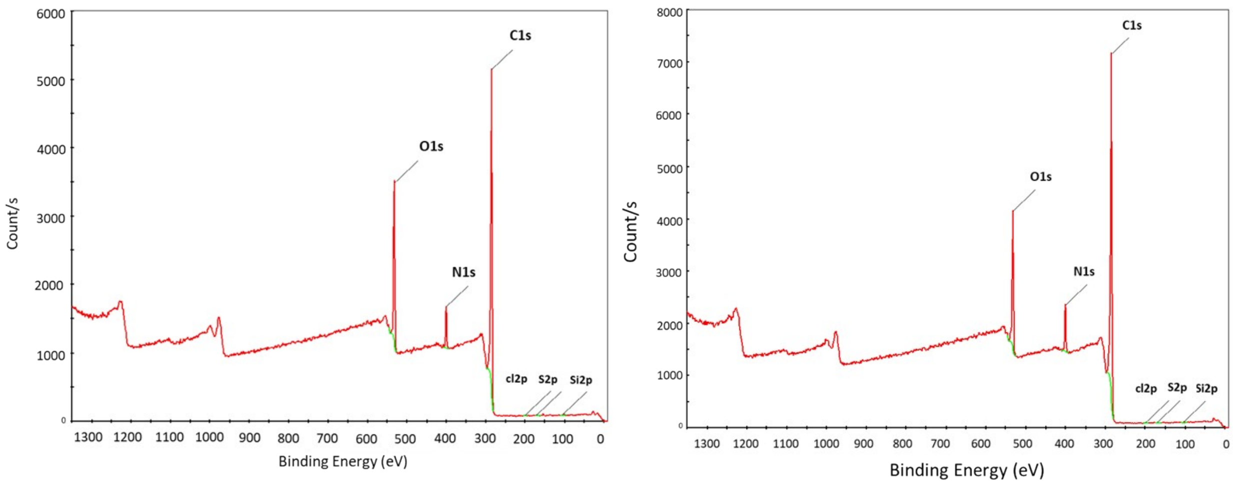


Figure S2. Histograms of fiber diameter measurement from SEM morphological graphs of PI aerogel samples

Table S2. Summary of average fiber diameters and standard deviations measured from SEM morphological graphs of PI aerogel samples

Aerogel Samples	**S1M**	**S1MF-M Skin**	**S1MF-M CR**	**S1MF-B Skin**	**S1MF-B CR**	**S2M**	**S2MF-M Skin**	**S2MF-M CR**	**S2MF-B Skin**	**S2MF-B CR**
Average Fiber Diameter (μm)	0.031	0.032	0.029	0.037	0.030	0.033	0.033	0.025	0.034	0.028
Standard Deviation	0.008	0.008	0.005	0.007	0.007	0.008	0.009	0.006	0.007	0.006



(a) (b)

Table S3. Relative atomic percentage of elements in PI aerogels

Sample	Relative Atomic % of Elements					
	Si 2p	Cl 2p	S 2p	C 1s	O 1s	N 1s
S1M	0.4	0.0	0.0	77.5	16.3	5.7
S2M	0.1	0.0	0.0	79.6	13.6	6.7

Figure S3. Low-resolution spectra of XPS data in (a) S1M and (b) S2M

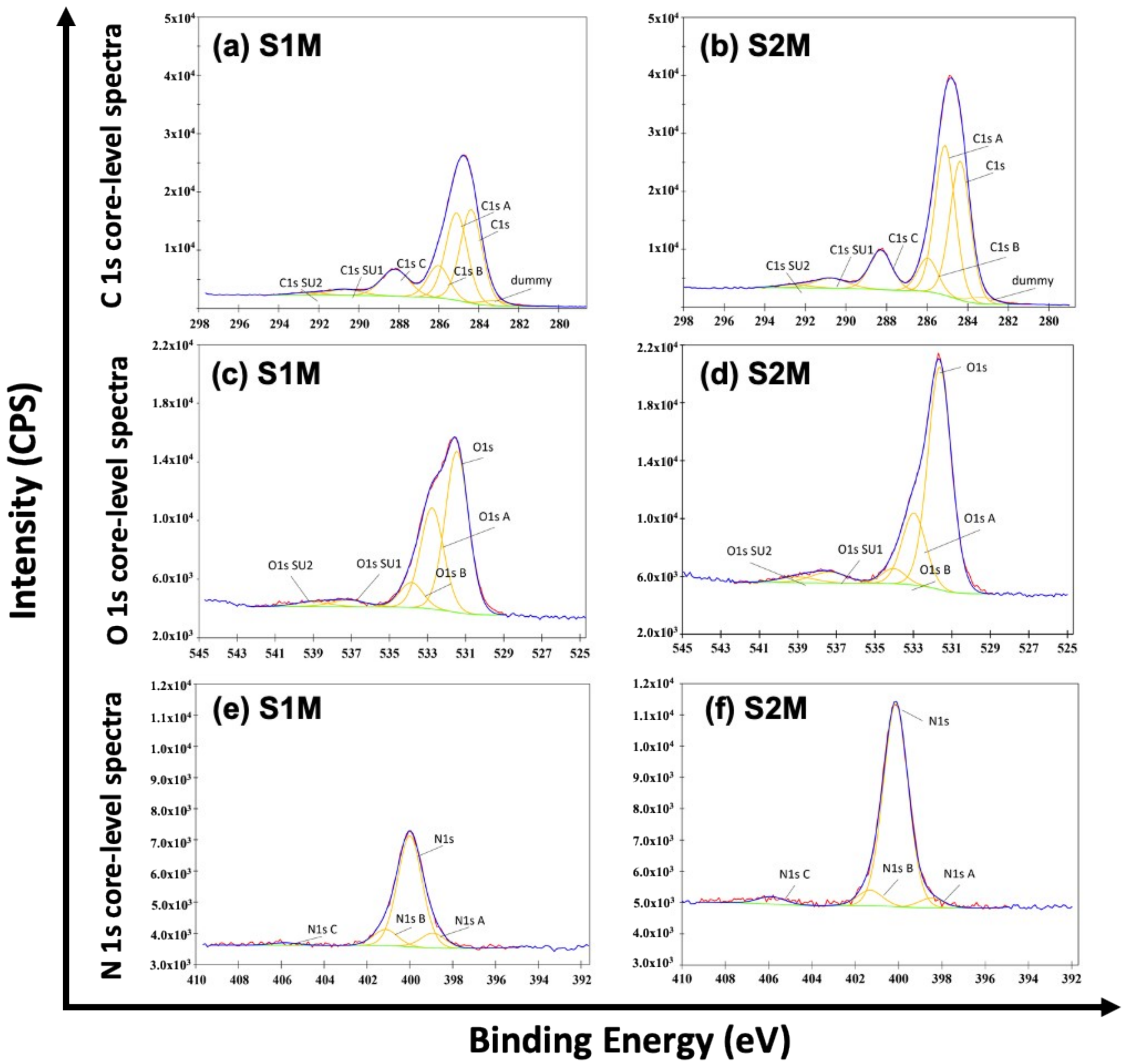


Figure S4. The XPS spectra decomposed by the Lorentzian-Gaussian fitting for the aerogel samples: (a) C 1s spectra of S1M; (b) C 1s spectra of S2M; (c) O 1s spectra for S1M; (d) O 1s spectra for S2M; (e) N 1s spectra for S1M; (f) N 1s spectra for S2M

Figure S5. Flammability test of aerogels (S1M on the top and S2M in the middle) compared with the one for natural wood (on the bottom)

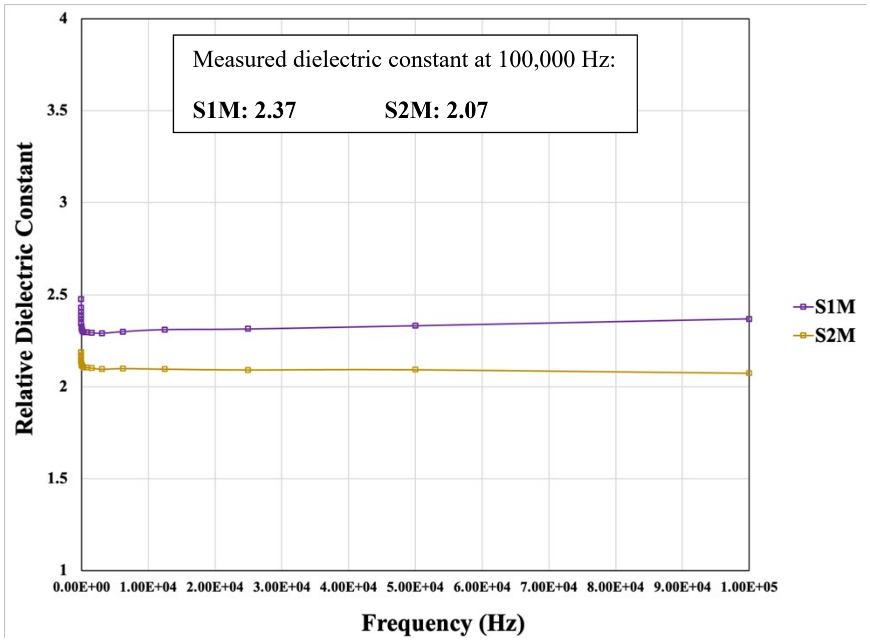


Figure S6. Relative dielectric constant measurement of PI aerogel samples in 0-10^5 Hz frequency

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