Experiment-in-Loop Interactive Optimization of Polymer Composites for "5G-and-Beyond"

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SI Tables

Table S1 Detailed information of the filler used in the experiment.

Filler producer	Filler name	Filler structure	Surface chemistry
ACG	Sunlovely	Length: 4－6um（thickness<0.1μm）plate filler	-/Perfluorohexylethyl Triethoxysilane
Kowa	KSF-3N	diameter: 7.5μm, filament filler	-/Triethoxy(pentafluorophenyl)silane
Kowa	TKV	diameter: 0.7～2.1 μm, filament filler	-/Triethoxy(pentafluorophenyl)silane
Nipponsteel	HS-208	diameter: 20 μm, Spherical filler	-/Triethoxy(pentafluorophenyl)silane
Nipponsteel	HS-311	diameter: 2.2 μm, Spherical filler	-/Triethoxy(pentafluorophenyl)silane

Table S2 Dataset collected in BO process.

Sample No.	Sample properties					Fabrication parameters							
	CTE (ppm/K)	ε	tan σ	Extinction coefficient	FOM	Time (min)	Rotation (rpm)	Plate filler condition	Spherical filler condition	Filament filler condition	Weight of plate filler (g)	Weight of spherical filler (g)	Weight of filament filler (g)
0	98.9	2.44	3.E-03	4.E-03	-0.110	7	100	1	3	2	2	3	2
0	67.0	2.62	3.E-03	5.E-03	0.000	9	200	1	0	3	2	2	3
0	76.7	2.42	3.E-03	5.E-03	-0.010	7	150	1	0	0	2	0	2
0	121.3	2.26	6.E-03	8.E-03	-0.803	5	50	1	0	3	5	0	1
0	52.2	2.55	3.E-03	5.E-03	0.102	5	250	1	0	2	2	0	5
0	73.0	2.52	1.E-03	2.E-03	0.323	7	250	0	1	1	1	4	2
1	38.8	2.44	6.E-04	9.E-04	0.691	5	250	0	1	0	0	2	4
1	95.7	2.42	1.E-03	2.E-03	0.181	5	250	0	0	0	1	6	0
1	66.4	2.37	7.E-04	1.E-03	0.492	9	250	0	3	0	0	1	5
1	112.5	2.32	8.E-04	1.E-03	0.160	5	250	0	0	3	0	2	1
1	122.3	2.25	6.E-04	9.E-04	0.141	5	250	0	3	0	0	7	0
2	35.9	2.41	6.E-04	1.E-03	0.706	5	250	0	0	0	0	0	7
2	67.4	2.39	6.E-04	1.E-03	0.495	5	50	1	3	0	0	2	5
2	61.9	2.50	6.E-04	1.E-03	0.533	5	250	0	0	0	0	2	5
2	49.9	2.43	8.E-04	1.E-03	0.572	5	250	0	3	0	0	1	6
2	32.7	2.44	7.E-04	1.E-03	0.716	5	50	0	2	0	0	2	5
3	40.2	2.52	7.E-04	1.E-03	0.653	5	100	0	1	0	0	1	6
3	32.9	2.43	6.E-04	9.E-04	0.738	9	50	0	0	0	0	0	7
3	31.8	2.53	9.E-04	1.E-03	0.683	5	50	0	1	0	0	1	6
3	28.3	2.38	9.E-04	1.E-03	0.704	5	50	0	0	0	0	0	7
3	47.9	2.49	6.E-04	9.E-04	0.628	7	150	0	1	0	0	1	6
4	24.8	2.56	6.E-04	1.E-03	0.782	7	50	0	0	0	0	0	7
4	40.2	2.49	7.E-04	1.E-03	0.652	7	100	0	0	0	0	0	7
4	28.8	2.52	2.E-03	2.E-03	0.577	9	100	0	0	0	0	0	7
4	28.5	2.49	6.E-04	1.E-03	0.750	5	100	0	0	0	0	0	7
4	43.6	2.01	2.E-03	3.E-03	0.389	9	50	0	0	3	0	0	7
5	94.7	2.18	6.E-04	9.E-04	0.327	5	50	0	0	0	0	0	3
5	44.4	2.33	7.E-04	1.E-03	0.633	5	50	0	0	0	0	0	6
5	109.9	2.14	6.E-04	9.E-04	0.223	7	50	0	0	0	0	0	3
5	57.8	2.46	6.E-04	1.E-03	0.557	5	50	0	0	0	1	0	6
5	82.7	2.22	6.E-04	8.E-04	0.410	5	50	0	0	0	0	0	4
6	25.4	2.58	8.E-04	1.E-03	0.746	5	50	0	0	0	0	1	6
6	120.3	2.24	4.E-04	7.E-04	0.180	9	50	0	0	0	0	7	0

6	42.1	2.38	8.E-04	1.E-03	0.632	9	50	0	0	0	0	3	4
6	75.5	2.18	7.E-04	1.E-03	0.426	9	50	0	0	0	0	2	5
6	74.0	2.50	3.E-03	4.E-03	0.047	9	250	0	3	0	6	1	0
7	45.2	2.38	6.E-04	9.E-04	0.648	5	150	0	0	0	0	0	7
7	58.1	2.58	6.E-04	1.E-03	0.559	5	50	0	0	0	0	4	3
7	38.8	2.66	7.E-04	1.E-03	0.658	5	50	0	0	0	0	3	4
7	40.5	2.56	7.E-04	1.E-03	0.649	5	50	0	1	0	0	4	3
7	93.1	2.27	7.E-04	1.E-03	0.310	5	250	0	3	0	0	1	2
8	49.8	2.48	7.E-04	1.E-03	0.603	5	250	0	1	0	0	1	6
8	61.8	2.41	9.E-04	1.E-03	0.482	5	250	0	2	0	0	1	6
8	57.9	2.53	8.E-04	1.E-03	0.527	7	250	0	1	0	0	1	6
8	51.7	2.34	9.E-04	1.E-03	0.548	5	250	0	2	0	0	1	6
8	58.4	2.67	2.E-03	3.E-03	0.258	9	50	0	3	3	0	1	6
9	71.4	2.61	2.E-03	3.E-03	0.165	5	150	0	3	3	0	1	6
9	83.9	2.56	2.E-03	4.E-03	0.036	9	250	0	3	3	0	3	4
9	94.4	2.56	2.E-03	3.E-03	0.102	5	250	0	3	3	0	3	4
9	82.0	2.59	2.E-03	3.E-03	0.124	5	250	0	0	3	0	0	7
9	69.7	2.61	3.E-03	4.E-03	0.082	9	250	1	3	3	1	1	5
10	24.9	2.65	1.E-03	2.E-03	0.669	5	50	0	0	1	0	0	7
10	86.9	2.22	1.E-03	2.E-03	0.275	5	250	0	0	0	0	0	3
10	36.3	2.59	1.E-03	2.E-03	0.597	7	50	0	0	1	0	0	7
10	56.6	2.45	1.E-03	2.E-03	0.403	5	150	1	3	0	1	1	5
10	35.5	2.58	2.E-03	2.E-03	0.527	5	50	1	0	0	1	0	6
11	113.2	2.28	7.E-04	1.E-03	0.172	9	50	0	0	3	0	6	1
11	77.7	2.57	3.E-03	5.E-03	-0.040	5	250	1	1	0	1	3	3
11	72.7	2.51	1.E-03	2.E-03	0.323	5	50	0	0	3	0	4	3
11	72.4	2.50	1.E-03	2.E-03	0.330	5	50	0	0	2	0	3	4
11	81.8	2.50	3.E-03	4.E-03	0.018	9	250	1	2	0	1	4	2

Table S3 CTE and dielectric performance compared with previous studies.

Ref.	Component	Filler dimensions	Filler diameter/shape	Filler (wt%)	CTE (ppm/K)	ε	tanδ	Extinction coefficient	FOM
This study	SiO_2/PTFE	0.7-2.1 nm/ filament	pristine	0.46	24.76	2.56	6.00×10^{-4}	9.50×10^{-4}	0.782
Alhaji et al.[10]	SiO_2/PTFE	25 μm/ spherical	pristine	-	55.77	2.18	1.10×10^{-2}	1.62×10^{-2}	-1.336
	SiO_2/PTFE	106 μm/ spherical	pristine	-	64.86	2.07	1.00×10^{-3}	1.44×10^{-3}	0.453
Liu et al.[3]	SiO_2/PTFE	≤10 μm/ spherical	Methyltriethoxysilane	0.40	141.00	2.30	1.10×10^{-3}	1.67×10^{-3}	-0.085
Li et al.[39]	SiO_2/PTFE	20 μm/ spherical (hollow)	pristine	0.40	95.00	1.94	8.40×10^{-4}	1.17×10^{-3}	0.285
Jin et al.[7]	SiO_2/PTFE	- / spherical	γ-methacryloxypropyltrimethoxysilane + pentafluorostyrene	0.30	86.00	2.38	1.00×10^{-3}	1.54×10^{-3}	0.299
Chen et al.[40]	SiO_2/PTFE	25 μm/ spherical	Phenyltrimethoxy silane	0.60	33.20	2.97	2.50×10^{-3}	4.31×10^{-3}	0.306
Chen et al.[11]	SiO_2/PTFE	5 μm/ spherical	Phenyltrimethoxy silane	0.60	40.00	2.80	1.50×10^{-3}	2.51×10^{-3}	0.486
Yuan et al.[8]	SiO_2/PTFE	9 μm/ spherical	Phenyltrimethoxy silane	0.57	32.30	2.89	7.00×10^{-4}	1.19×10^{-3}	0.702
Jiang. et al.[7]	SiO_2/PTFE	2 μm + 15 μm/ spherical (mixed)	Phenyltrimethoxysilane/Aminopropyltriethoxysilane	0.62	18.60	2.99	2.00×10^{-3}	3.46×10^{-3}	0.510
Zheng et al.[5]	CLST/PTFE	-	-	0.40	45.00	7.92	1.20×10^{-3}	3.38×10^{-3}	0.344
Subodh et al.[4]	PTFE/TeO_2	-	tetrabutyltitanate	0.60	32.00	5.40	6.00×10^{-3}	1.39×10^{-2}	-0.890
Wang et al.[6]	PTFE/BCZN	0.456 μm /near-spherical	Perfluorooctyltriethoxysilane	0.50	33.00	7.70	1.40×10^{-3}	3.88×10^{-3}	0.361
Huang et al.[31]	epoxy resin /BNNT	50 nm /nanotube	oligosilsesquioxane	0.30	53.00	3.50	2.50×10^{-2}	4.68×10^{-2}	-5.134
Sasikala et al.[29]	PS/SiO_2	0.4 μm /spherical	3-Aminopropyltriethoxysilane/vinyltriethoxysilane	0.11	50.00	3.15	1.60×10^{-2}	2.84×10^{-2}	-2.817
Xue et al.[30]	SiO_2–PEEK	7 μm+10 μm+20μm/ Spherical (mixed)	3-Aminopropyltriethoxysilane	0.60	23.60	3.35	1.63×10^{-3}	2.98×10^{-3}	0.536

Table S4 Structural character of Sample #A~#E.

Sample No.	Density (g/cm^3)	Intensity @788cm^{-1}	Intensity @2367cm^{-1}	Amorphous index (from FTIR)	Amorphous degree (from XRD)
#A	2.15	8.60×10^{-2}	3.15×10^{-3}	27.30	0.581
#B	2.15	5.90×10^{-2}	1.04×10^{-2}	5.65	0.616
#C	2.13	1.19×10^{-1}	9.90×10^{-2}	12.02	0.630
#D	2.06	1.62×10^{-1}	1.26×10^{-2}	12.90	0.655
#E	2.08	1.47×10^{-1}	1.30×10^{-2}	11.27	0.657

Table S5 Parameters for EMT calculations

G_p (GPa)	G_v (GPa)	G_f (MPa)	k_p (GPa)	k_v (GPa)	k_f (GPa)	a_p (ppm/K)	a_f (ppm/K)	v_f
0.24	0	0.42	1.36	0	36.3	159	0.5	0.46

SI Figures

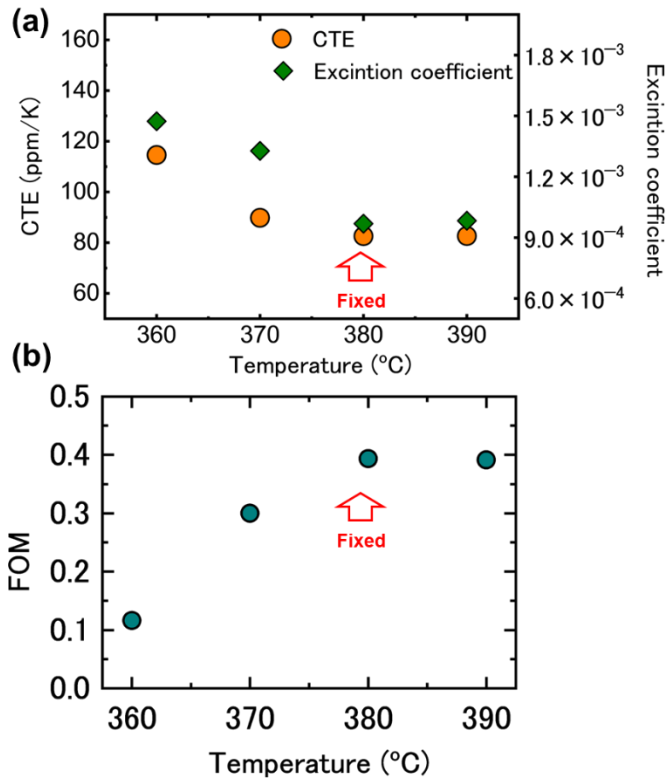

Figure S1. The performance of the silica/PFA under different compounding temperature. (a) the CTE and the dielectric extinction coefficient, (b) the FOM of PFA/silica composite compounded under different temperature. The composites consist of the filament filler of 3.5 g, and PFA of 11.5 g; the rotation speed is 50 rpm. (When compounding temperature exceed 370 ºC, the FOM achieve a acceptable value and saturate.

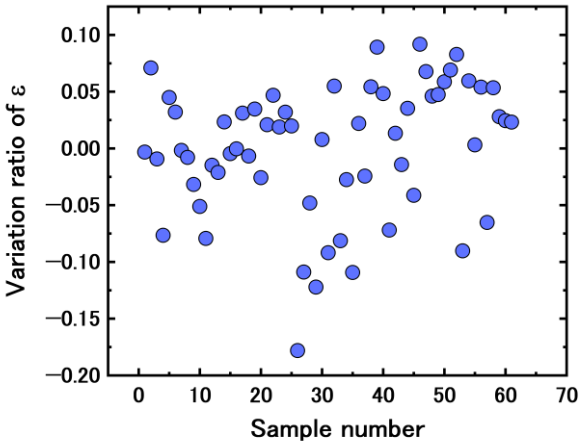

Figure S2. The variation ratio of ε during the BO process.

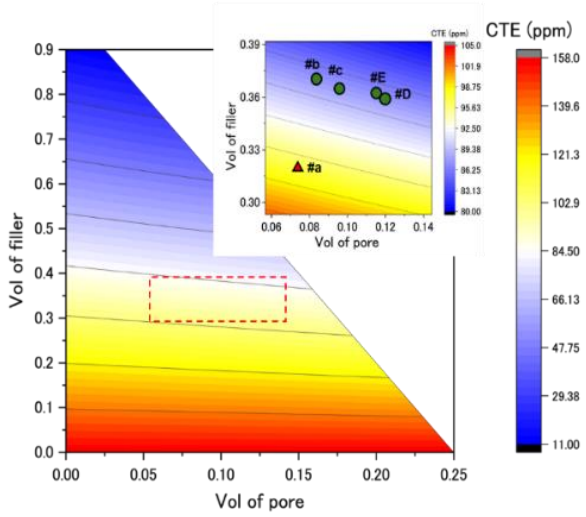

Figure S3. EMT calculation for the CTE of composite. Theoretical CTE values of a three-phase composite consist of PFA, silica, and pore, calculated by EMT with the parameters obtained for samples #A~#E.

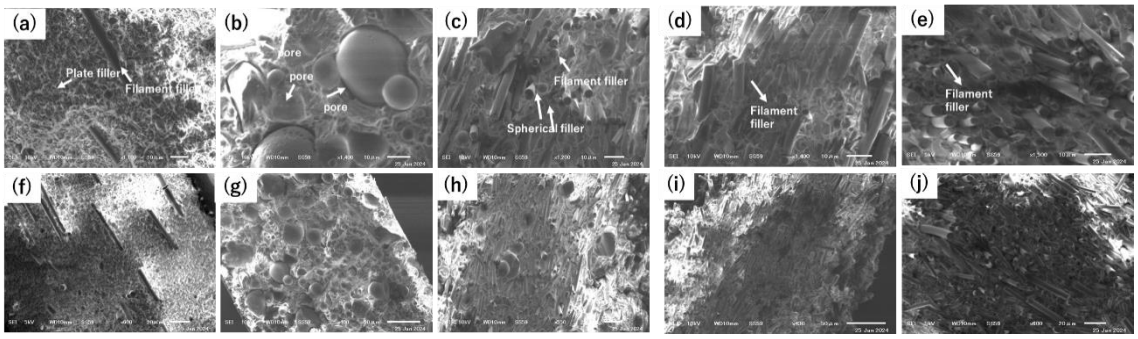

Figure S4. SEM image of PFA/silica composite. Cross-section SEM image of the Sample (a)(f) #A, (b)(g) #B, (c)(h) #C, (d)(i) #D, (e)(j) #E under (a)-(e) low and (f)-(j) high magnification.

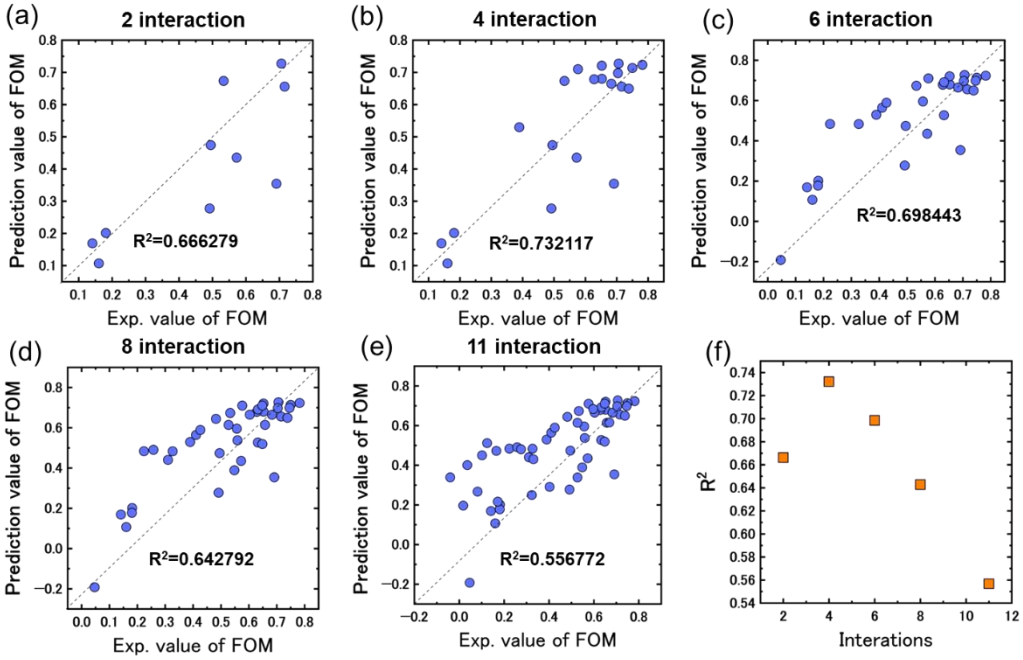

Figure S5. Parity plots of BO. (a)-(e) Parity plots of FOM during the BO under an interaction number of 2, 4, 6, 8, 11, respectively; (f) Variation of R^2 under different interactions.