

Appendix. Supplementary Information

# **Preparation of bottom-up graphene oxide using citric acid and tannic acid, and its application as a filler for polypropylene nanocomposites**

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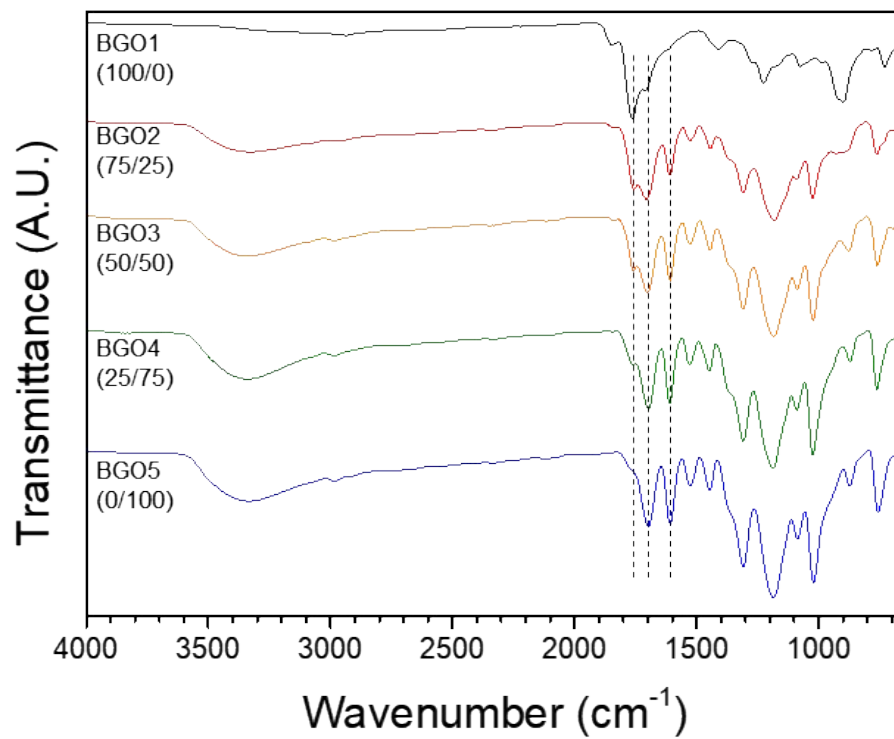
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Figure S1 FT-IR spectrum of BGOs prepared by different weight ratio of CA/TA.



**Figure S2** TEM images of (a) BGO1, (b) BGO2, (c) BGO3, (d) BGO4 and (e) BGO5.

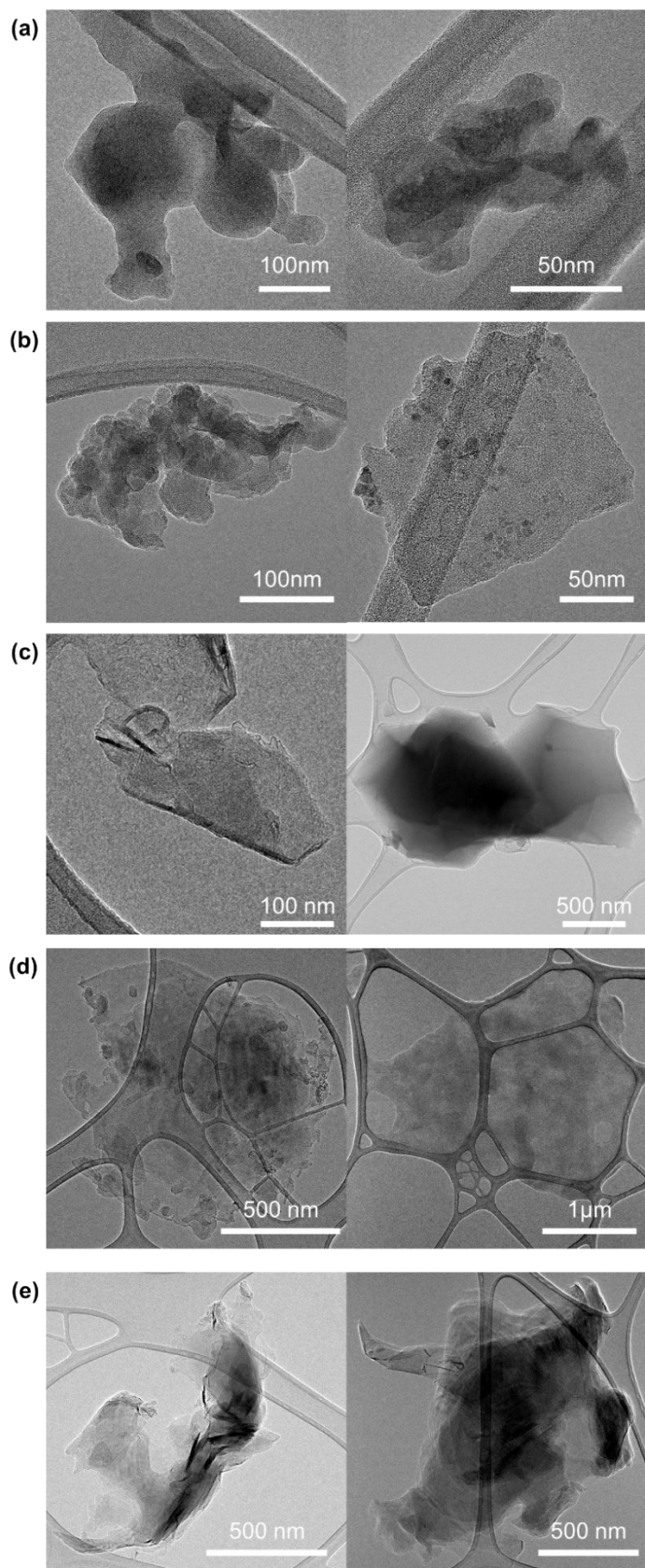
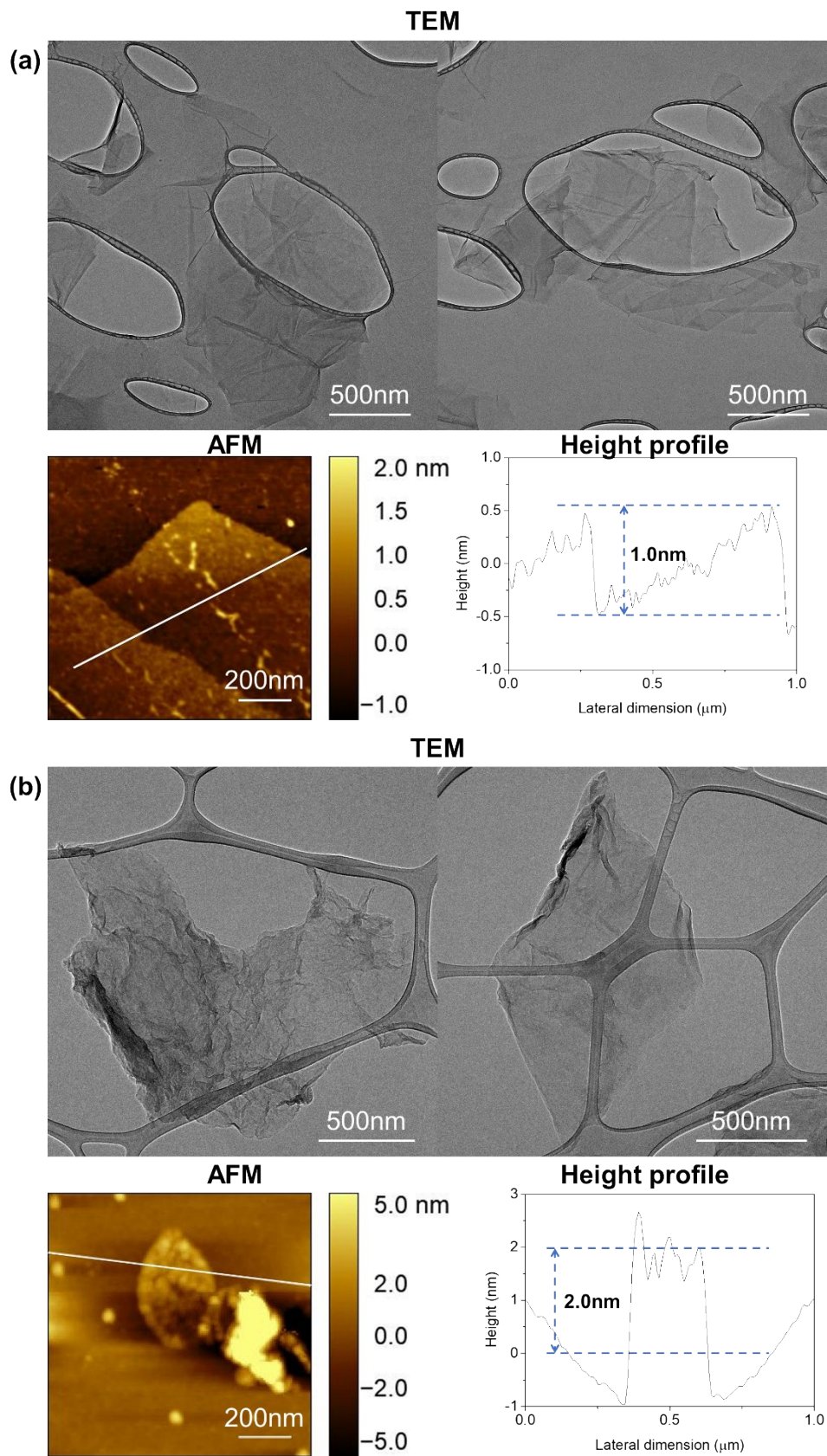
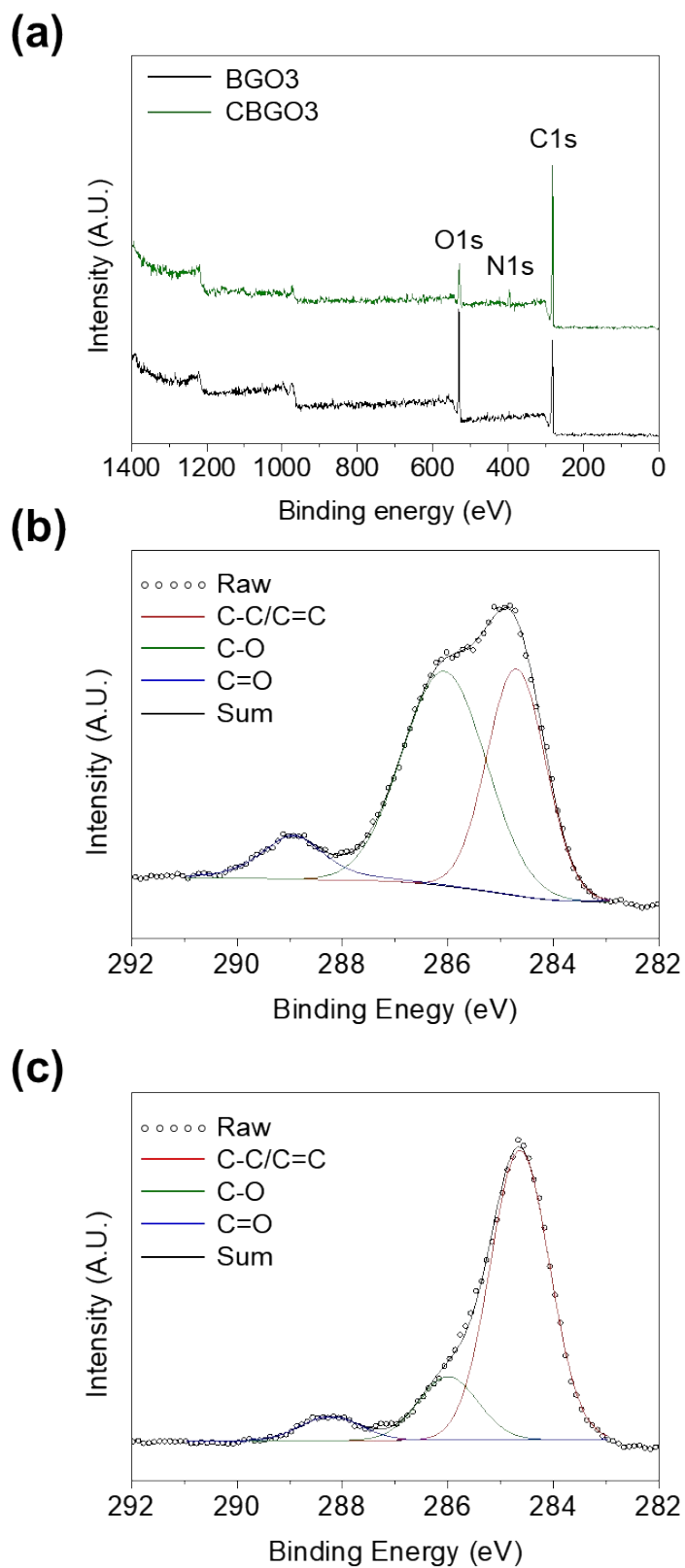


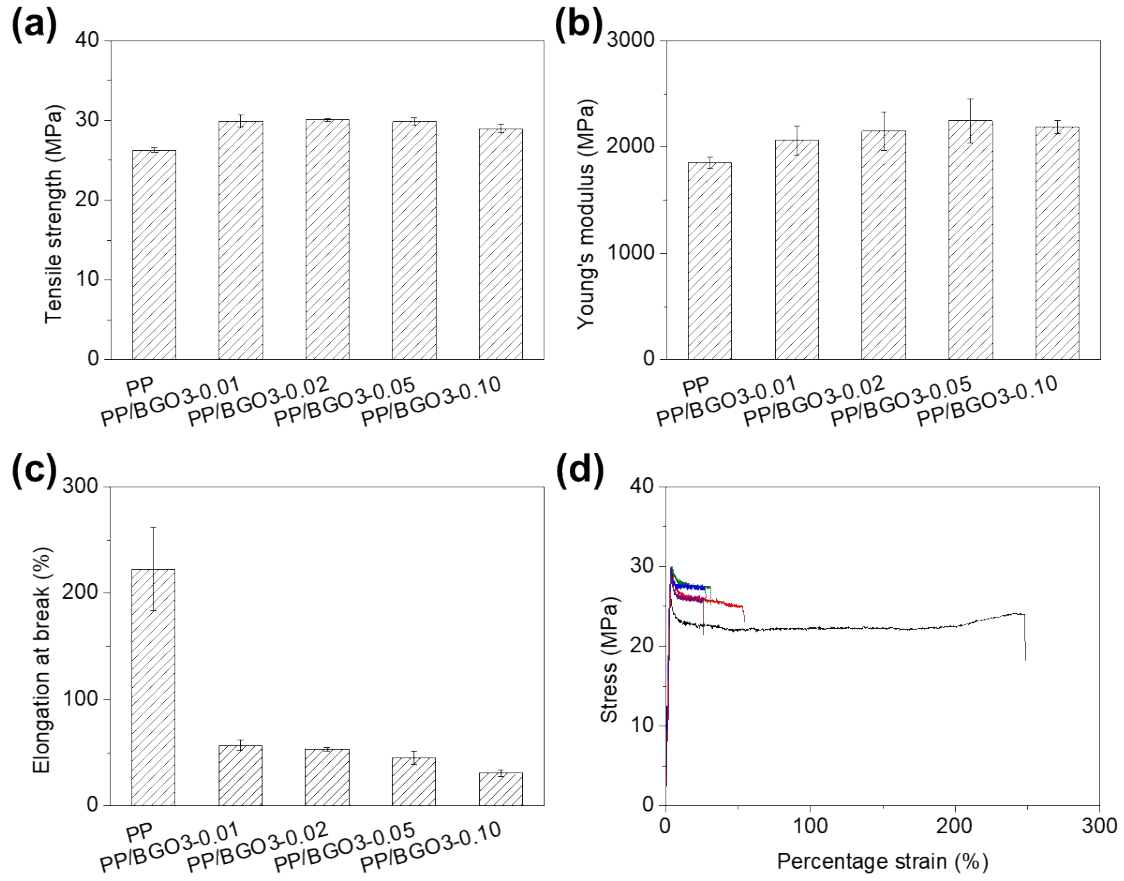
Figure S3 TEM images, AFM image, and height profile of (a) GO and (b) CGO.



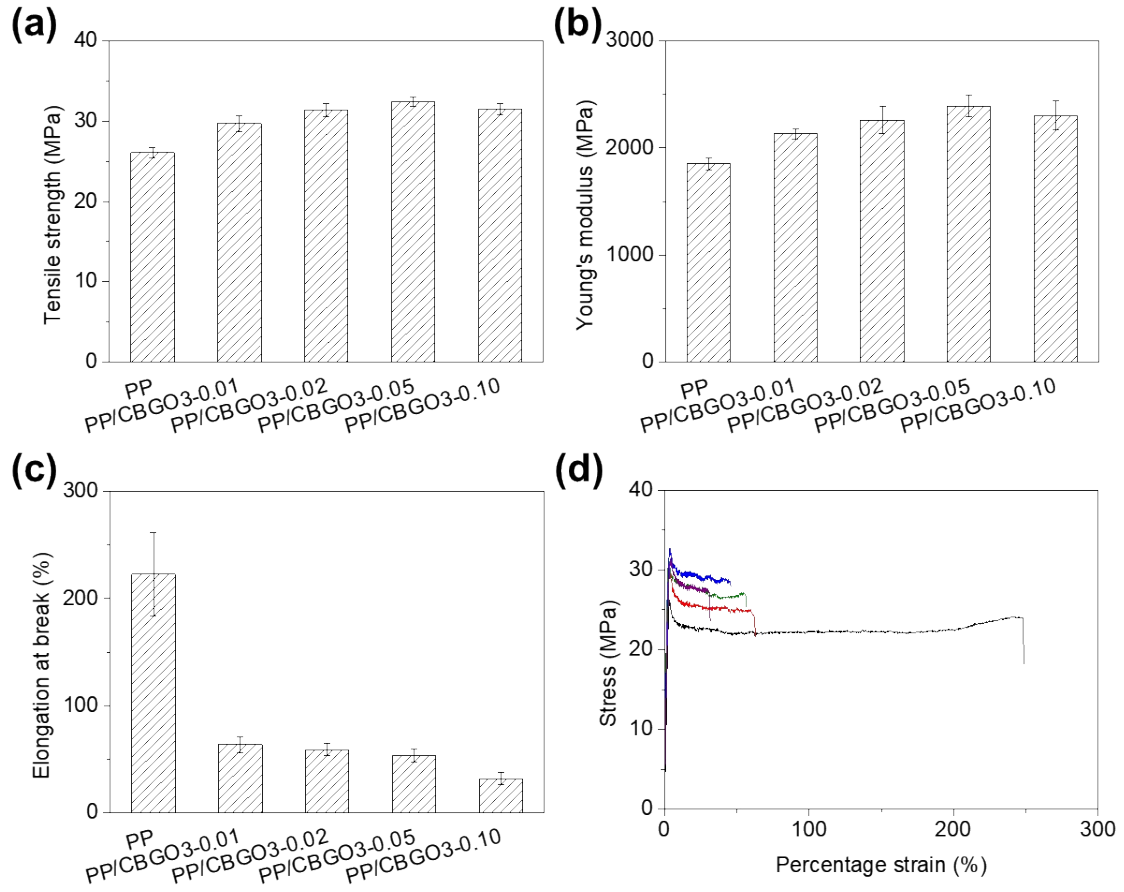
**Figure S4** (a) XPS wide scan spectrum of BGO3 and CBGO3, (b) XPS C1s scan of BGO3, and (c) XPS C1s scan of CBGO3.



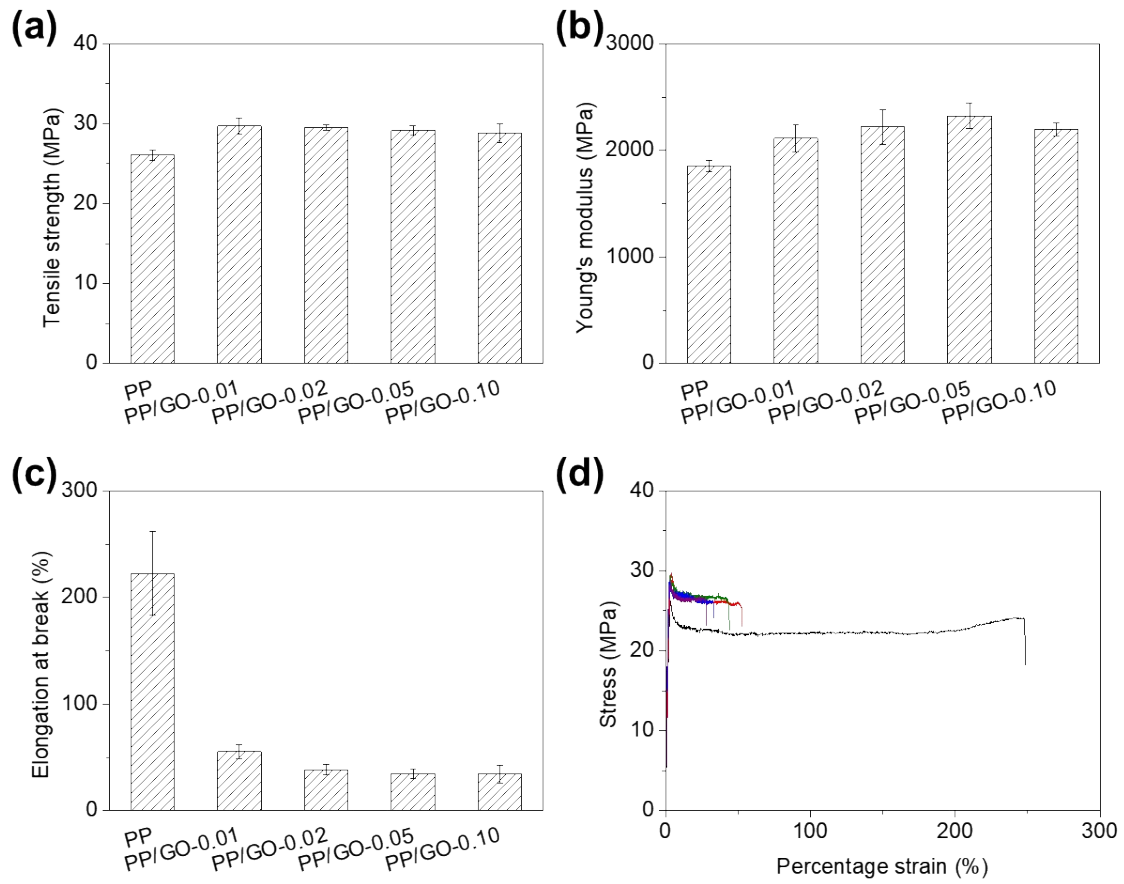
**Figure S5** Tensile test results of PP/BGO3 nanocomposites. (a) Tensile strength, (b) Young's modulus, (c) elongation at break, and (d) representative strain-stress curves.



**Figure S6** Tensile test results of PP/CBGO3 nanocomposites. (a) Tensile strength, (b) Young's modulus, (c) elongation at break, and (d) representative strain-stress curves.

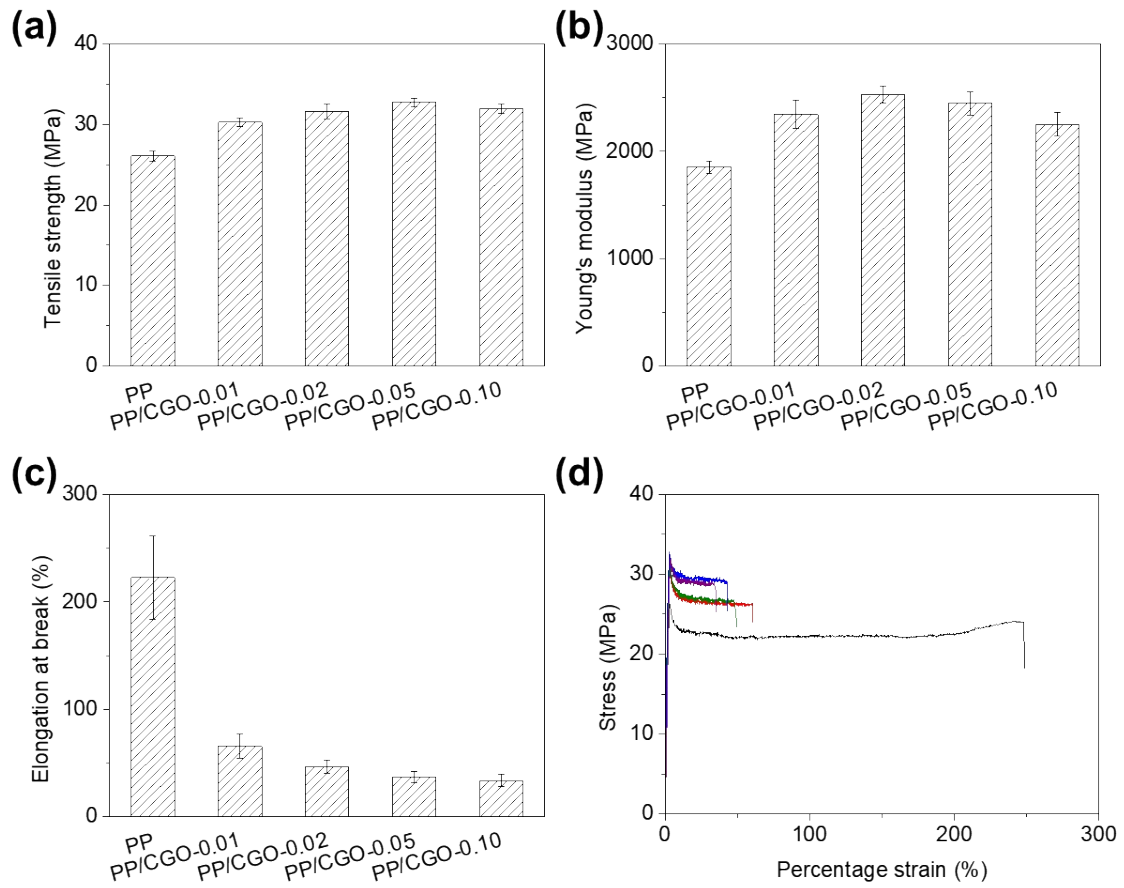


**Figure S7** Tensile test results of PP/GO nanocomposites. (a) Tensile strength, (b) Young's modulus, (c) elongation at break, and (d) representative strain-stress curves.

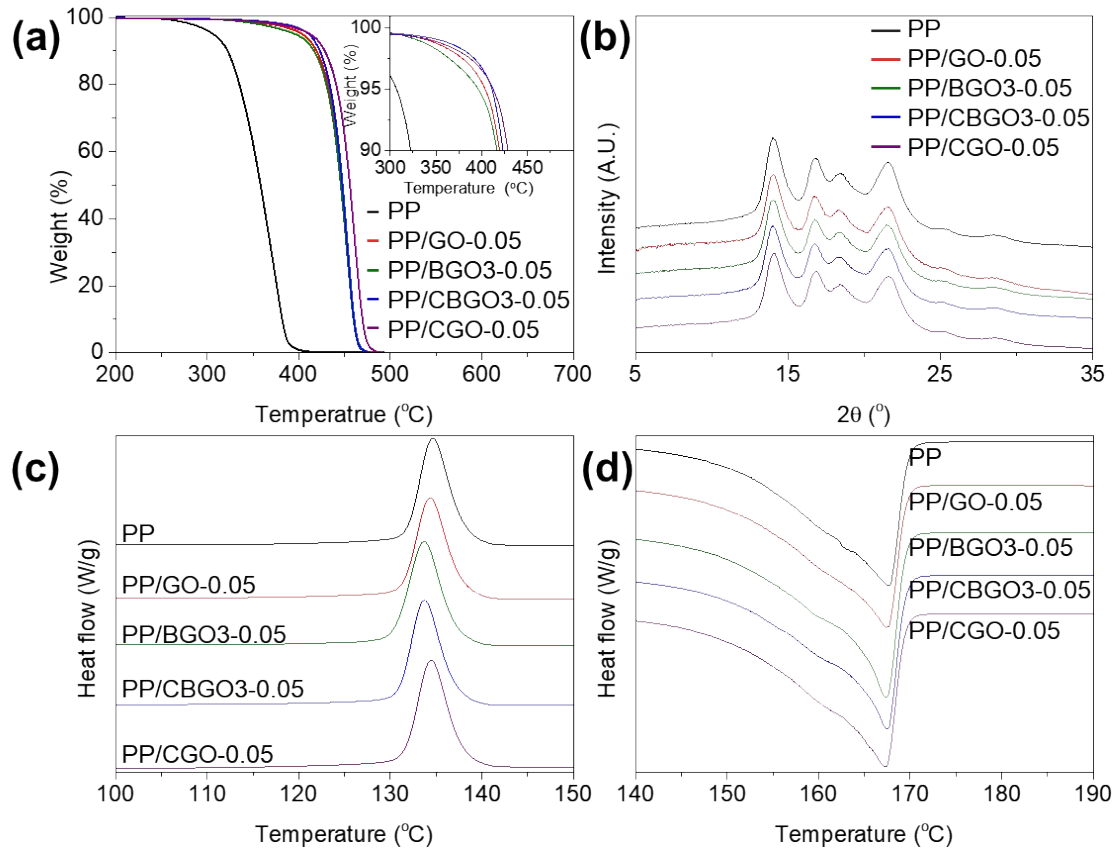




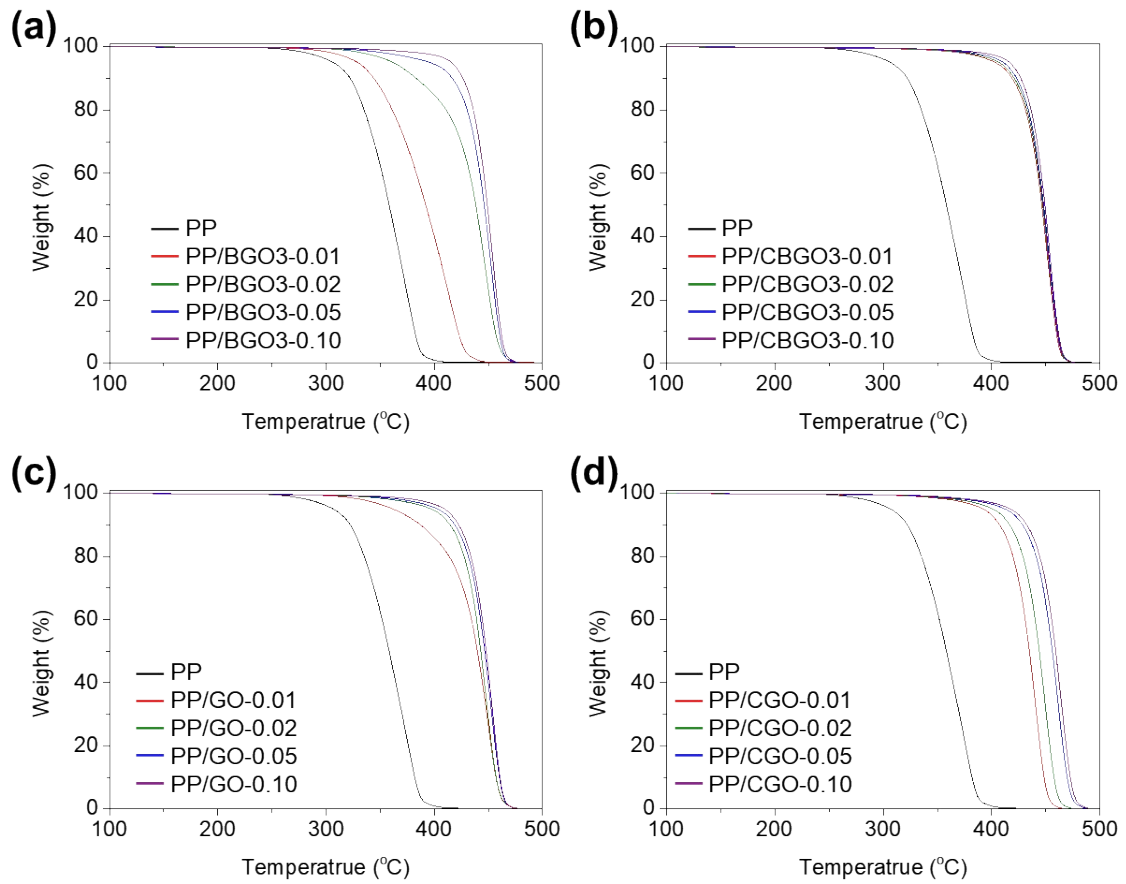
**Figure S8** Tensile test results of PP/CGO nanocomposites. (a) Tensile strength, (b) Young's modulus, (c) elongation at break, and (d) representative strain-stress curves.



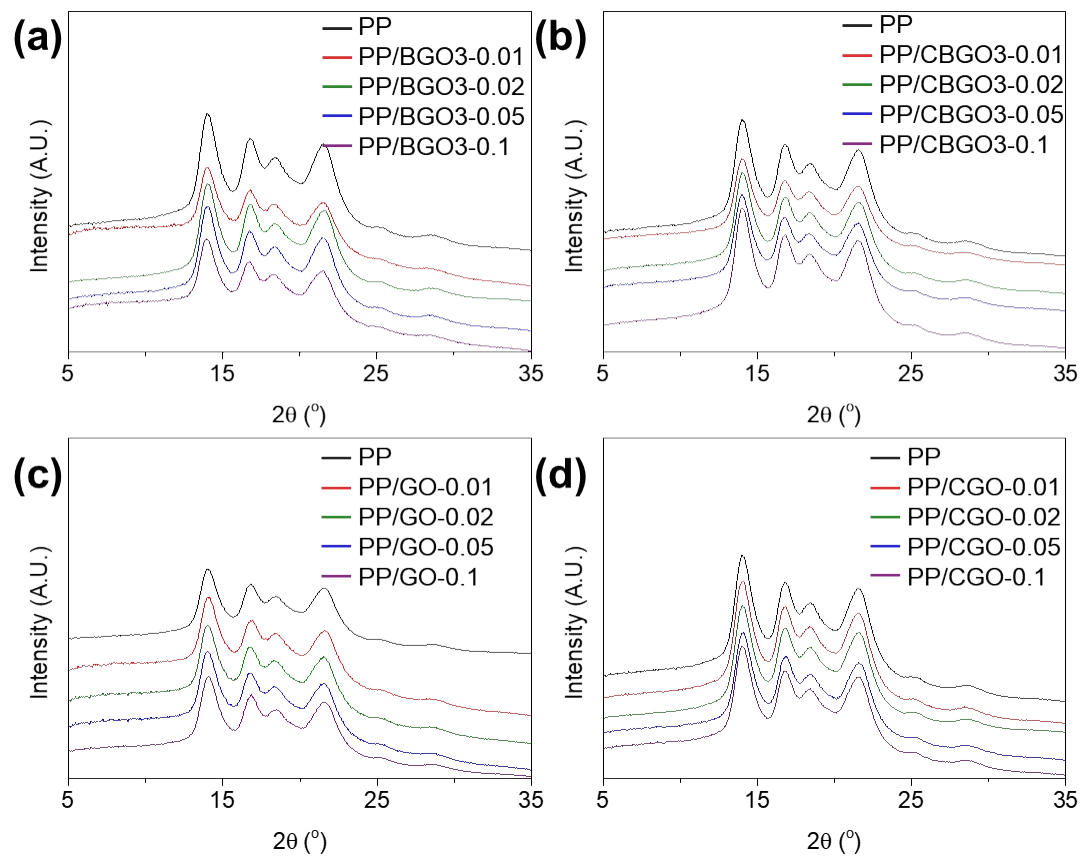
**Figure S9** (a) TGA curves, (b) XRD patterns, (c) DSC cooling curves, and (d) DSC heating curves of pristine PP and PP nanocomposites containing 0.05 wt% of fillers.



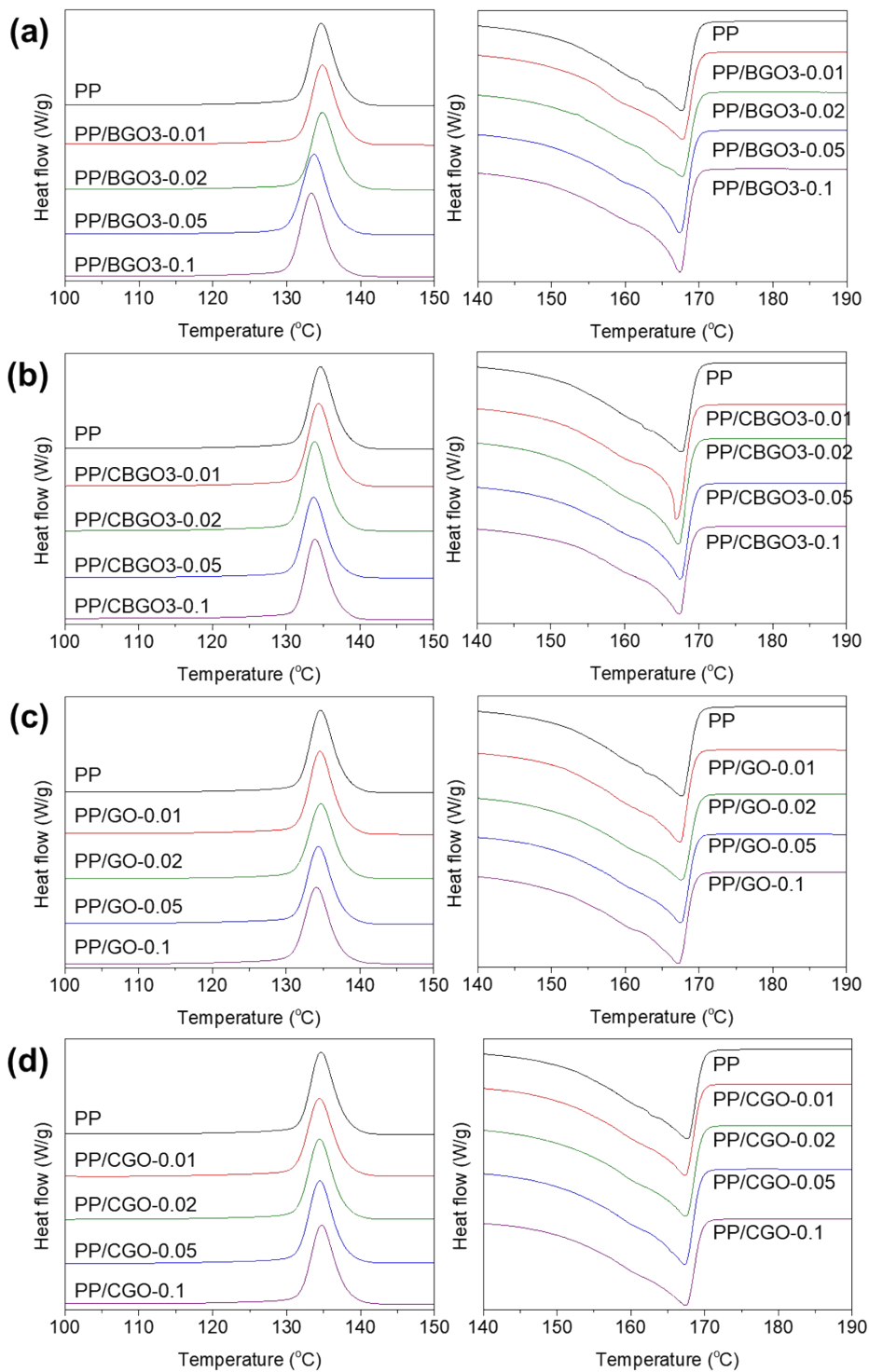
**Figure S10** TGA curves of PP nanocomposites. (a) PP/BGO3, (b) PP/CBGO3, (c) PP/GO and (d) PP/CGO nanocomposites.



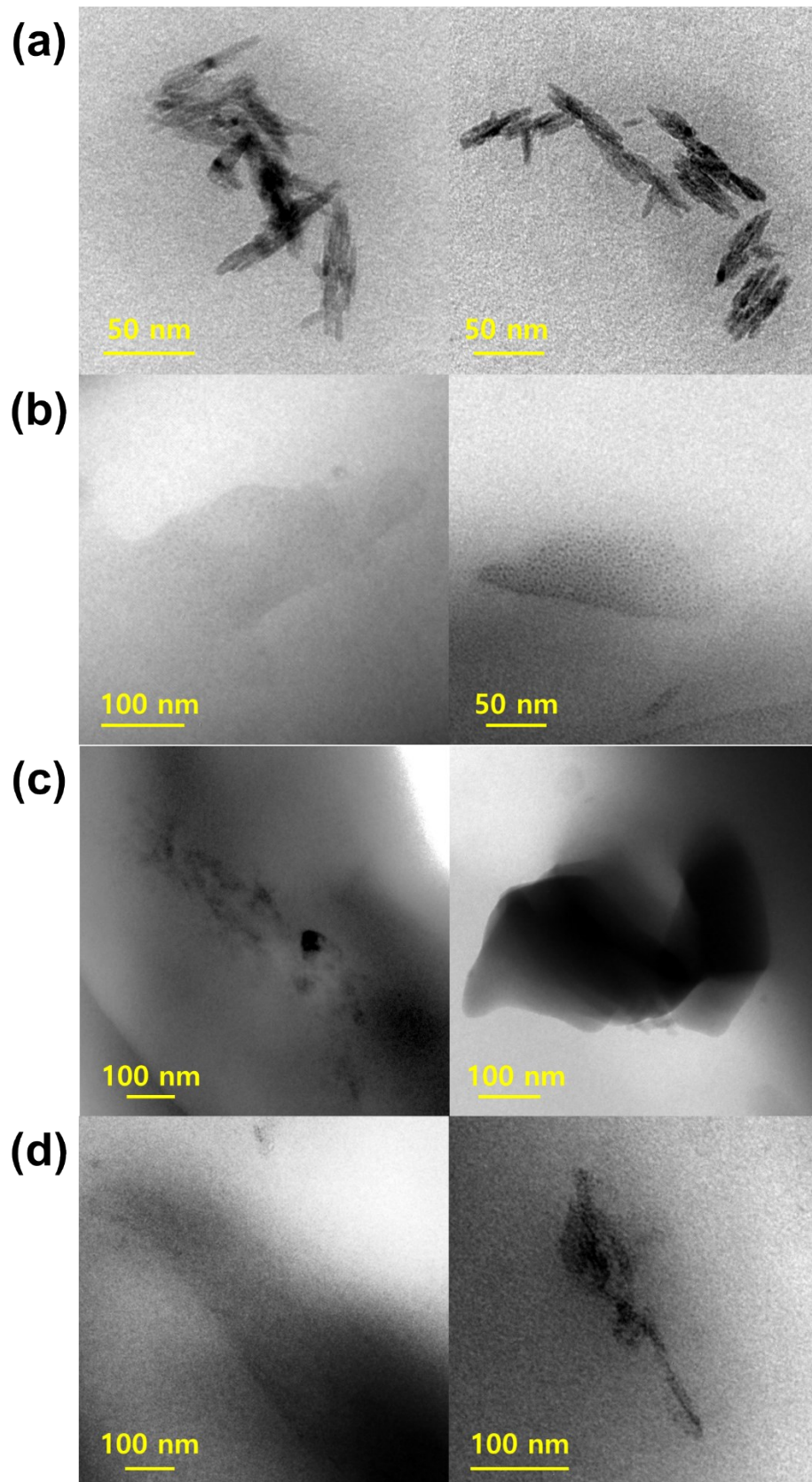
**Figure S 11** XRD patterns of PP nanocomposites. (a) PP/BGO, (b) PP/CBGO3, (c) PP/GO, and (d) PP/CGO nanocomposites.



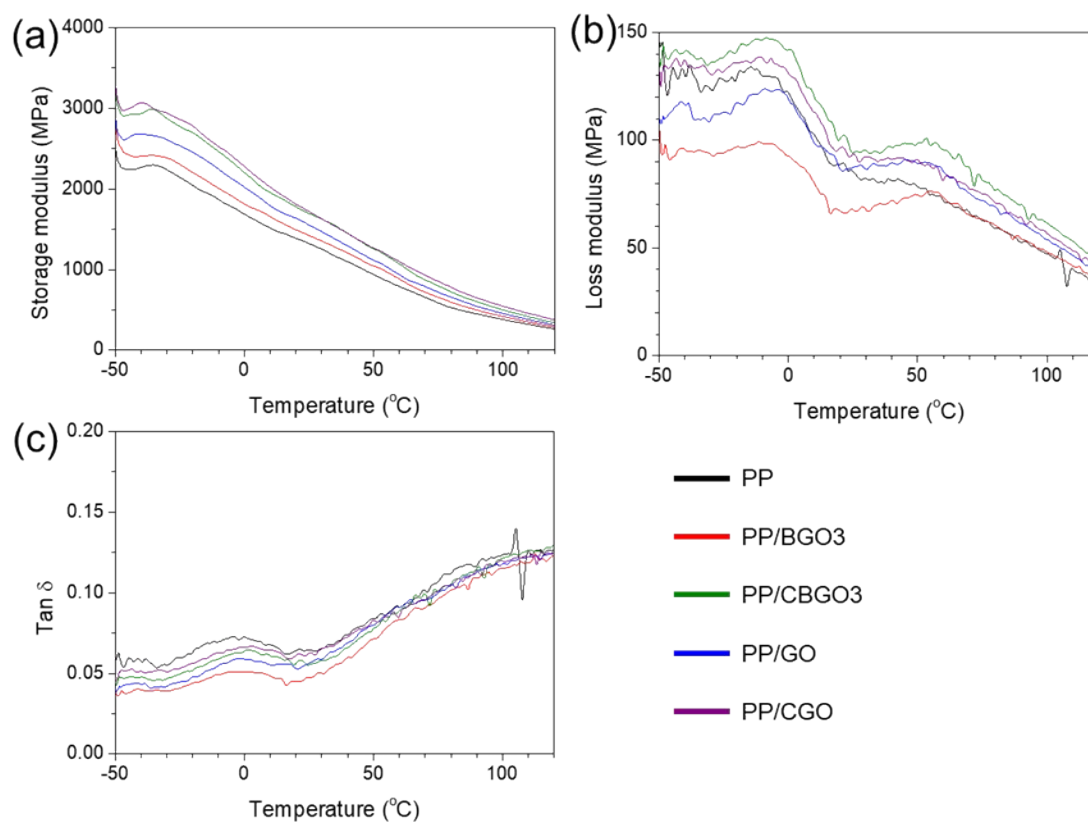
**Figure S 12** DSC results of PP nanocomposites. (a) PP/BGO3 nanocomposites, (b) PP/CBGO3 nanocomposites, (c) PP/GO nanocomposites, and (d) PP/CGO nanocomposites.



**Figure S13** TEM images of fillers in PP nanocomposites containing 0.05 wt% of fillers. (a) BGO3. (b) CBGO3. (c) GO. (d) CGO.



**Figure S 14** DMA results of pristine PP and PP nanocomposites. (a) Storage modulus values. (b) Loss modulus values. (c) Tan  $\delta$  values.



**Table S1** Elemental analysis result of BGOs and CBGO3.

Sample	CA/TA <sup>a</sup>	Content (wt%)			
		C	H	O	N
BGO1	100/0	53.3	4.1	42.5	0.0
BGO2	75/25	55.9	4.0	40.1	0.0
BGO3	50/50	51.7	3.9	44.4	0.0
BGO4	25/75	52.8	4.2	43.0	0.0
BGO5	0/100	51.4	3.9	44.7	0.0
CBGO3	-	55.2	4.8	39.4	0.6

<sup>a</sup> The weight ratio of CA/TA used for the preparation of BGO.



**Table S 2** Mechanical properties and Thermal degradation temperature for 5 wt% loss ( $T_{d,5}$ ) of pristine PP and PP nanocomposites.

Sample	Tensile strength (MPa)	Young's modulus (MPa)	Elongation at break (%)	$T_{d,5}$ (°C)
PP	26.1±0.3	1853.4±57.1	222.7±38.8	307.0
PP/BGO3-0.01	29.9±0.7	2061.0±137.6	56.9±4.6	329.7
PP/BGO3-0.02	30.1±0.1	2152.2±180.1	53.2±1.8	362.4
PP/BGO3-0.05	29.9±0.5	2247.5±208.4	45.0±6.3	396.4
PP/BGO3-0.10	29.0±0.5	2186.2±62.1	30.7±3.2	416.3
PP/CBGO3-0.01	29.7±1.0	2132.3±49.7	63.6±7.5	403.6
PP/CBGO3-0.02	31.4±0.8	2260.6±124.6	58.8±5.6	407.2
PP/CBGO3-0.05	32.4±0.6	2392.5±103.6	53.2±6.4	411.2
PP/CBGO3-0.10	31.5±0.7	2302.3±136.4	31.7±5.5	416.9
PP/GO-0.01	29.7±1.0	2113.8±130.6	55.5±6.6	362.0
PP/GO-0.02	29.5±0.4	2219.3±166.8	38.3±4.8	397.4
PP/GO-0.05	29.1±0.6	2323.7±119.4	34.7±4.0	403.3
PP/GO-0.10	28.8±1.2	2199.3±60.6	34.4±8.2	409.7
PP/CGO-0.01	30.3±0.5	2344.5±133.1	65.4±11.6	392.5
PP/CGO-0.02	31.6±1.0	2526.1±78.4	46.3±5.9	401.2
PP/CGO-0.05	32.8±0.5	2448.2±109.7	36.3±5.4	412.7
PP/CGO-0.10	32.0±0.5	2251.6±107.0	33.3±5.5	417.6

**Table S 3** DSC results of pristine PP and PP nanocomposites.

Sample	Cooling			Heating		
	Onset temperature	Peak temperature	Heat of crystallization	Onset temperature	Peak temperature	Heat of melting
	(°C)	(°C)	(J/g)	(°C)	(°C)	(J/g)
PP	138.2	134.7	92.1	152.8	167.6	87.3
PP/BGO3-0.01	138.3	134.8	87.6	154.4	167.7	86.2
PP/BGO3-0.02	138.3	134.9	86.0	152.9	167.6	84.0
PP/BGO3-0.05	137.3	133.7	92.5	159.8	167.3	89.2
PP/BGO3-0.1	136.8	133.4	90.3	160.4	167.4	87.8
PP/CBGO3-0.01	137.9	134.4	92.6	165.1	166.9	87.2
PP/CBGO3-0.02	137.4	133.9	98.8	158.9	167.2	94.5
PP/CBGO3-0.05	137.2	133.8	89.1	159.5	167.4	84.0
PP/CBGO3-0.1	137.4	133.9	83.8	158.3	167.4	77.2
PP/GO-0.01	138.1	134.6	91.8	155.7	167.4	86.7
PP/GO-0.02	138.2	134.7	89.2	153.2	167.6	86.7
PP/GO-0.05	137.9	134.4	89.0	156.3	167.3	84.7
PP/GO-0.1	137.6	134.1	88.1	155.7	167.1	87.6
PP/CGO-0.01	138.1	134.4	91.3	155.3	167.3	88.3
PP/CGO-0.02	137.9	134.4	91.0	155.0	167.4	87.6
PP/CGO-0.05	138.0	134.5	93.5	155.4	167.3	88.5
PP/CGO-0.1	138.2	134.7	87.0	153.8	167.3	83.9