Electronic Supplementary Information

Unexpected Segmental Dynamics in Polystyrene-Grafted Silica Nanocomposites

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Table S1 Formulations of silica NP-filled PS and PMMA nanocomposites

Sample code	Polymer matrix	Silica NP type	Silica NP content[a]
PS/SiO_2	PS	Bare	5 wt%
PS/SiO_2-PS	PS	PS grafted	5 wt%
PMMA/SiO_2	PMMA	Bare	5 wt%
PMMA/SiO_2-PS	PMMA	PS grafted	5 wt%

[a]For SiO_2-PS-filled nanocomposites, the silica NP content is excluding grafted PS chains.

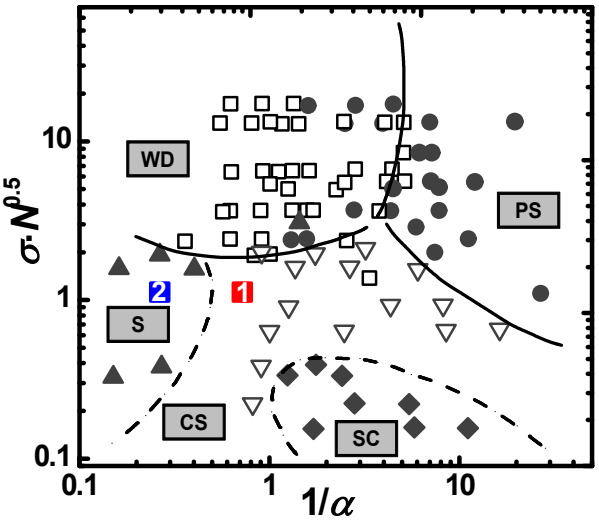

Fig. S1 Classical composite morphology diagram created based from ref. 13: $\sigma \cdot N^{0.5}$ as a function of $1/\alpha$ with $\alpha = N/P$. Points, adapted from the literature, are coded symbol, wherein open square symbols (□) correspond to well-dispersed particles (WD); solid circle symbols (●), phase separated samples (PS); solid triangle symbols (▲), strings (S); open triangle symbols (▽), connected sheets (CS); and solid diamond symbols (◆), small clusters (SC). Number-labeled color symbols 1 and 2 represent the respective spatial distribution of SiO_2-PS particles in PS and PMMA nanocomposites investigated in this study.

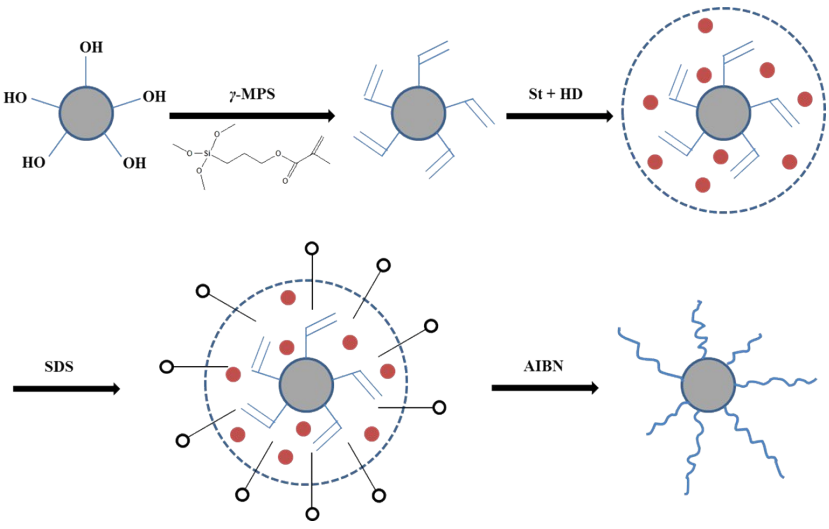

Fig. S2 Schematic for synthesis of SiO_2-PS composite particles via miniemulsion polymerization.

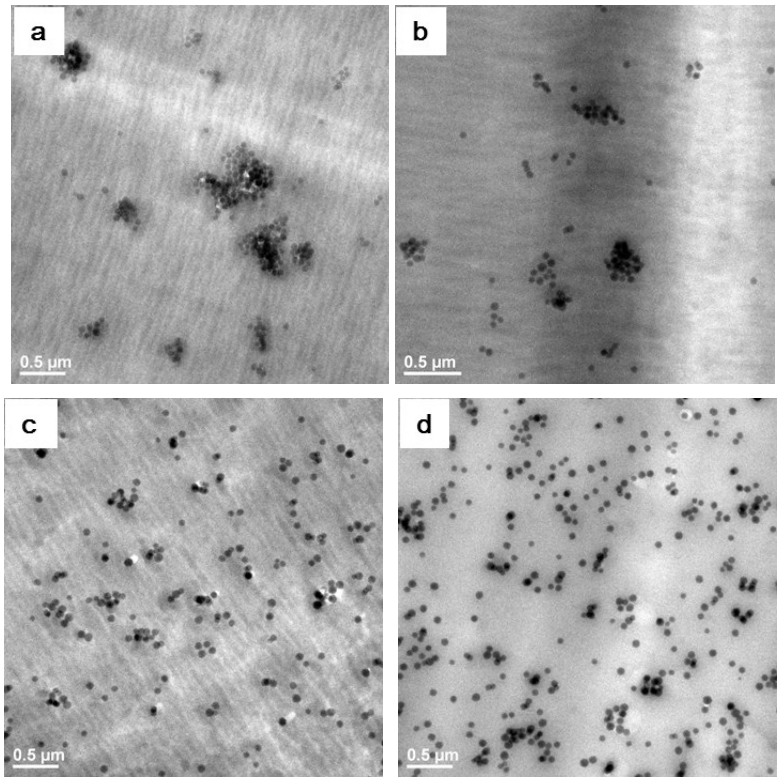

Fig. S3 TEM images of unmodified SiO_2-filled PS (a), PMMA (b), SiO_2-PS composite particle filled PS (c), and PMMA nanocomposites (d) after annealing at 423 K for 120 h. The content of silica NPs is 5 wt%.

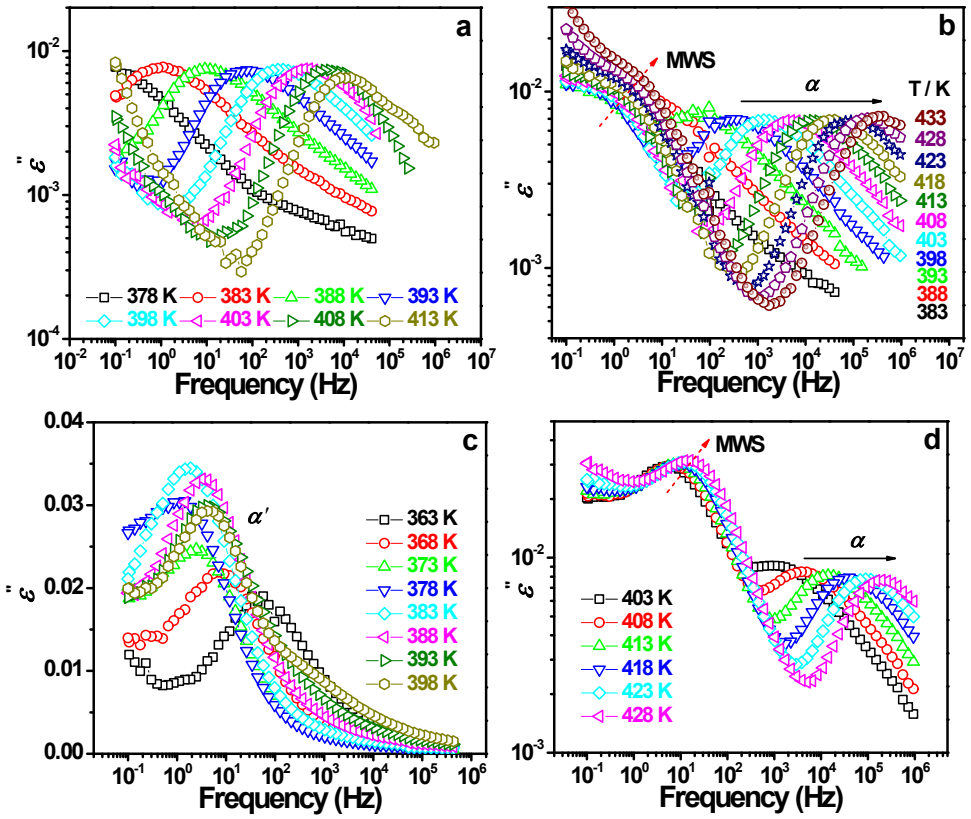

Fig. S4 Frequency dependence of dielectric loss for pure PS (a), SiO_2-filled PS (b) and SiO_2-PS-filled PS nanocomposites (c, d) at various temperatures.

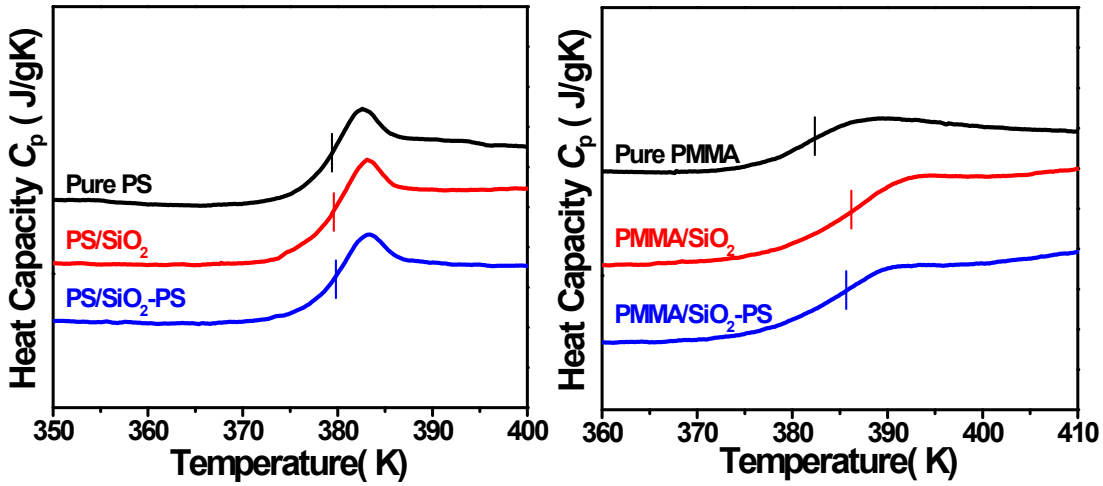

Fig. S5 DSC curves of pure PS and its nanocomposites (a), pure PMMA and its nanocomposites (b) at the heating rate of 10 K/min. The solid lines show the position of T_g.

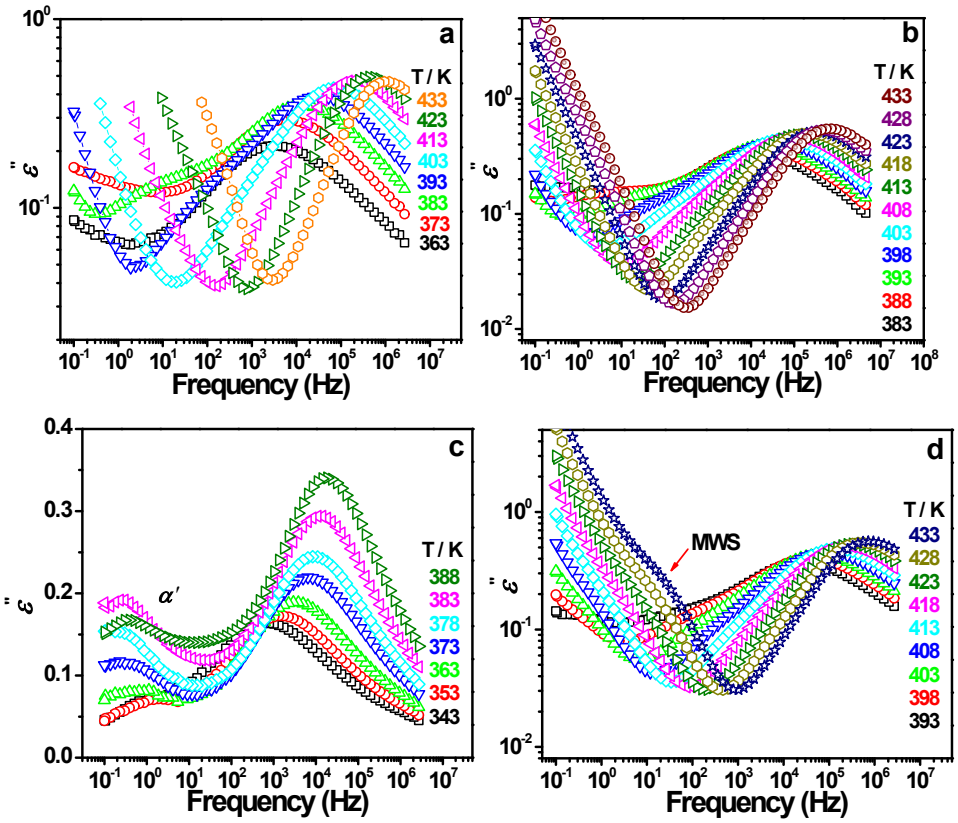

Fig. S6 Frequency dependence of dielectric loss for pure PMMA (a), SiO_2-filled PMMA (b) and SiO_2-PS-filled PMMA nanocomposites (c, d) at various temperatures.

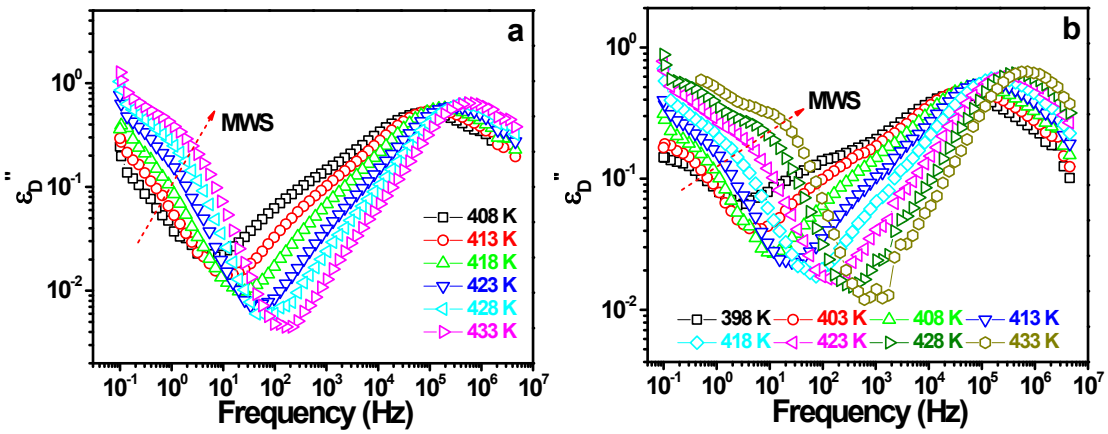

Fig. S7 Derivative spectra of PMMA nanocomposites filled with SiO_2 (a) and SiO_2-PS composite particles (b) at various temperatures.