Polyimide nanocomposites with functionalized SiO$_2$ nanoparticles: Enhanced processability, thermal and mechanical properties

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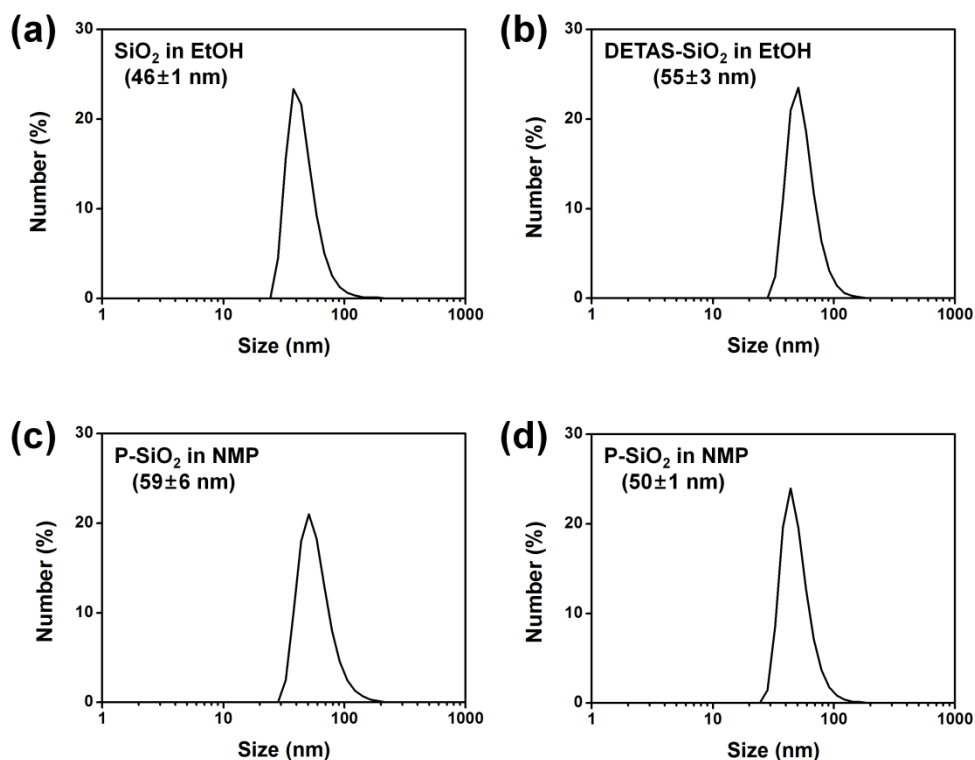[c]Present address: Department of Medicine, Emory University, Atlanta, GA 30322, USA

Figure. S1 Size distributions of (a) SiO$_2$ in EtOH, (b) DETAS-SiO$_2$ in EtOH, (c) as-prepared P-SiO$_2$ in NMP, and (d) P-SiO$_2$ in NMP after being stored for one year.
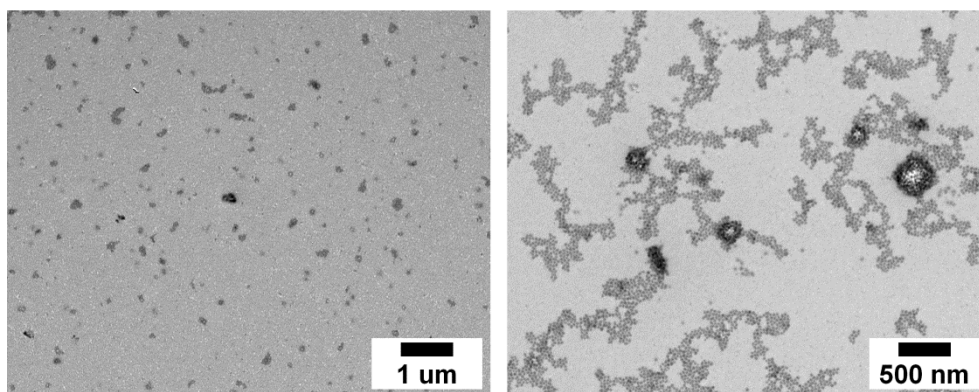
Figure. S2 TEM images of unmodified SiO_2-PI nanocomposites. The TEM images were obtained by spin-coating SiO_2-PAA solution onto a TEM grid, followed by thermal imidization.
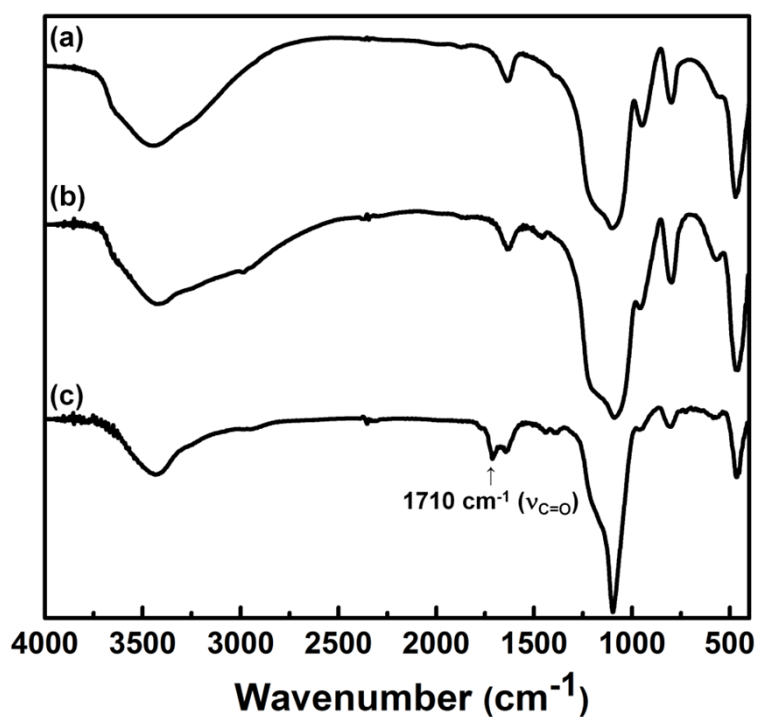

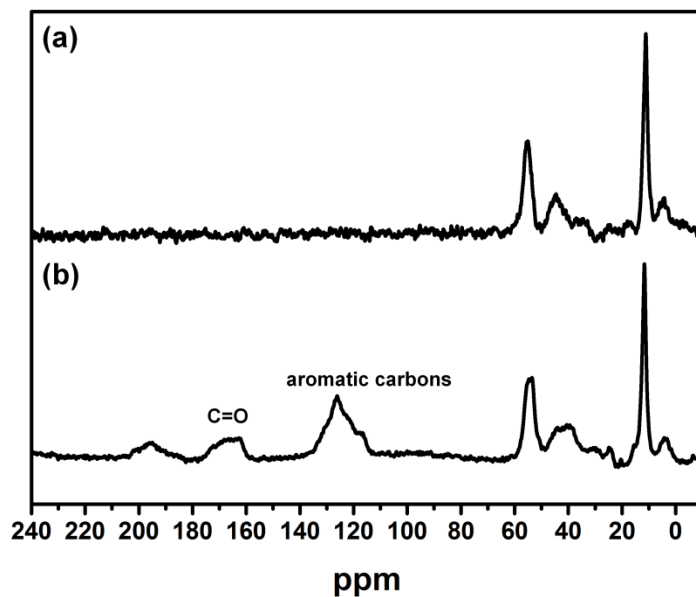**Figure. S3** FT-IR spectra of (a) unmodified silica nanoparticles, (b) DETAS-SiO_2, and (c) P-SiO_2.

Figure. S4 Solid ^{13}C CP/MAS NMR spectra of (a) DETAS-SiO$_2$ and (b) P-SiO$_2$.

Table S1. Calculated molar ratio of organic groups on SiO$_2$ nanoparticles by TGA.

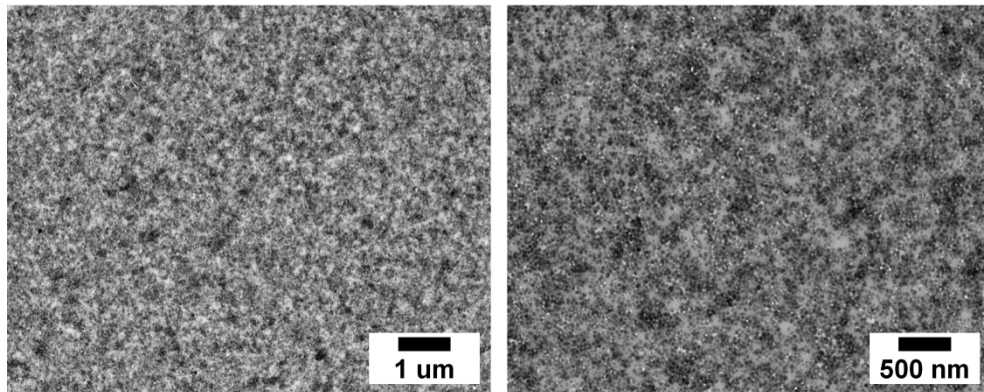|	Weight percent (wt%)	Molecular weight (g/mol)	Relative amount from 100 g sample (mol)	Mole percent of organic groups (mol%)
SiO$_2$	87.4	60 (for fully condensed SiO$_2$)	1.5	
DETAS on the DETAS-SiO$_2$	5.6	265.43 (for DETAS)	2.1×10^{-2}	1.4
Phthalic anhydride on the P-SiO$_2$	7	148.12 (for phtalic anhydride)	4.7×10^{-2}	3.1

Figure. S5 TEM images of 40 wt% SiO_2-PI nanocomposites. The TEM images were obtained by spin-coating SiO_2-PAA solution onto a TEM grid, followed by thermal imidization.
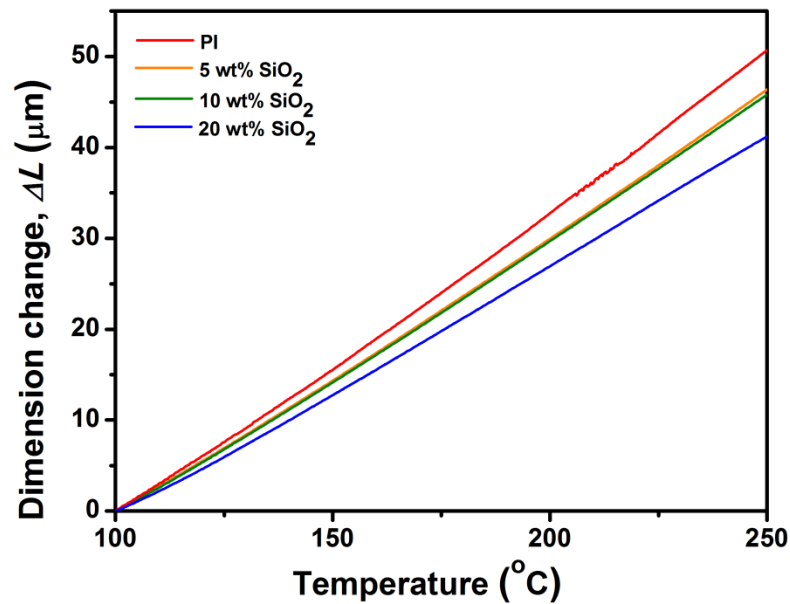

Figure. S6 Thermal expansion of neat PI film and SiO_2-PI nanocomposite films with various SiO_2 contents.

Calculation details

The mathematical equation[1] is described as follows:

$$\alpha_c = \alpha_f \phi_f + \alpha_m \phi_m$$

where the subscripts c, m, f represent nanocomposites, polymer, and filler phase, respectively. α and ϕ are coefficient of thermal expansion and the volume fraction of the constituents. The coefficient of thermal expansion for polyimide and SiO_2 are 33.9 and 0.55 (10^{-6}/°C), respectively.

Table S2. The experimental and calculated value of coefficients of thermal expansion of polyimide and SiO_2-polyimide nanocomposites

wt. %	vol. %	Experimental value (10^{-6}/°C)	Calculated value (10^{-6}/°C)
Neat PI	-	33.9	-
5	2.2	30.8	33.1
10	4.6	30.6	32.2
20	9.8	27.8	30.2

1.H. S. Katz and J. V. Mileski, *Handbook Of Fillers For Plastics*, Springer, 1987, 49.