Supplementary Materials: Radiation Induced Surface Modification of Nanoparticles and their Dispersion in Polymer Matrix

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$$w_{PVDF} = \frac{m_{graft\ PVDF}}{m_{F-SiO_2}}$$
$$= \frac{f_{SiO_2} \cdot (f_{SiO_2} - vinyl - f_{F-SiO_2})}{f_{SiO_2} \cdot f_{SiO_2} - vinyl \cdot (f_{F-SiO_2} - f_{PVDF}) + f_{SiO_2} \cdot (f_{SiO_2} - vinyl - f_{PVDF}) - f_{SiO_2} - vinyl \cdot (f_{F-SiO_2} - f_{PVDF})}$$

Equation S1. Equation for calculating the grafting content of PVDF on F-SiO₂ nanoparticles surface. Where w_{PVDF} is the grafting ratio of the grafted PVDF chain on the surface of SiO₂, which based on the TGA data. $m_{graft PVDF}$ is the actual number mass of grafting PVDF chain on the surface of SiO₂. m_{F-SiO_2} is the number mass of the F-SiO₂ nanoparticles, which calculated by the content of PVDF and γ -MPS on the F-SiO₂ nanoparticles surface. f_{SiO_2} , $f_{SiO_2-vinyl}$, f_{F-SiO_2} and f_{PVDF} were represented the inorganic residual content of pristine SiO₂, SiO₂-vinyl, F-SiO₂ and PVDF after the TGA test respectively.



Figure S1. ¹³C-NMR spectra of pristine SiO₂, SiO₂-vinyl and F-SiO₂ nanoparticles, respectively.

(1)



Figure S2. (a) XRD patterns of pristine SiO₂, SiO₂-vinyl, F-SiO₂ and PVDF; (b) Enlarged XRD patterns of pristine SiO₂, SiO₂-vinyl, F-SiO₂ in the 2 θ range of 15–30°. (c) Enlarged Rietveld refinement of the pristine SiO₂, SiO₂-vinyl and F-SiO₂ XRD data in the 2 θ range of 15–30°.



Figure S3. SEM and Elemental Mapping image (EMI) of PVDF matrix incorporated with 3wt% (**a**) pristine SiO₂, (**b**) SiO₂-vinyl and (**c**) F-SiO₂ nanoparticles, respectively; the subscript of 1 and 2 are correspond to the signal of silicon and fluoride element in EMI, respectively.



Figure S4. AFM of PVDF matrix incorporated with 3wt% (**a**) pristine SiO₂, (**b**) SiO₂-vinyl and (**c**) F-SiO₂ nanoparticles, respectively.



Figure S5. SEM image of PVDF matrix incorporated 5wt% (**a**), 20wt% (**b**), 35wt% (**c**) SiO₂ nanoparticles, respectively.



Figure S6. Digital photograph of PVDF matrix incorporated 3wt% (**a**–**g**) 35wt% SiO₂ nanoparticles, respectively; Digital photograph of PVDF matrix incorporated 3wt% (**h**–**n**) 35wt% F-SiO₂ nanoparticles, respectively.



Figure S7. DSC spectra of 1st cooling (**a**) and 2nd heating (**b**) of PVDF matrix incorporated with 3wt%, 5wt%, 20wt%, 35wt% SiO₂ nanoparticles, respectively.



Figure S8. XRD spectra of PVDF matrix incorporated with 3 wt% SiO₂, SiO₂-vinyl and F-SiO₂ nanoparticles, respectively.

Sample	d ^a (nm)	T _d ^b (°C)	f _{inorganic} b (%)	w _{γ-MPS} ^c (%)	W _{PVDF} ^c (%)
SiO ₂	10.3 ± 1.6	-	98.0	-	-
SiO ₂ -vinyl	10.1 ± 2.1	439.6	95.2	3.4	-
F-SiO ₂	15.3±3.5	467.4	74.5	3.4	35.9

^a Measured from TEM images (*d*: number averaged diameter of nanoparticles).^b Measured from the TGA curves of purified specimen (T_d : the TGA temperature with maximum loss rate; $f_{inorganic}$: inorganic residual content of modification nanoparticles after TGA test). ^c Calculated by equation (1) based on TGA data ($w_{\gamma-MPS}$: graft content of γ -MPS functioned on the silica nanoparticles surface; $w_{\gamma-MPS}$: graft content of PVDF grafted onto the silica nanoparticles surface;).

Table S2. The graft content of PVDF grafted onto silica nanoparticles under different reactant ratio (wt% /wt%) and irradiation dose.

Code	[SiO ₂]:[PVDF] (wt% / wt%)	Absorbed Dose (kGy)	Time (h)	T_d^a (°C)	finorganic b (%)	WPVDF ^b (%)
1	1:0.1	30	17	433.3	92.7	4.0
2	1:0.3	30	17	436.9	90.0	6.8
3	1:0.5	30	17	438.2	88.8	8.1
4	1:0.7	30	17	445.7	82.0	15.1
5	1:1	30	17	460.4	79.5	17.7
6	1:2	30	17	467.4	74.5	26.1
7	1:2	50	17	465.7	67.0	30.6
8	1:2	70	17	458.1	66.4	31.3

^a Measured from the TGA curves of purified specimen (T_d : the TGA temperature with maximum loss rate; $f_{inorganic}$: inorganic residual content of modification nanoparticles after TGA test).^b Calculated

by equation (1) based on TGA data ($w_{\gamma-MPS}$: graft content of PVDF grafted onto the silica nanoparticles surface).