Electronic Supplementary Information

1. DLS of the NPAM-silica emulsion

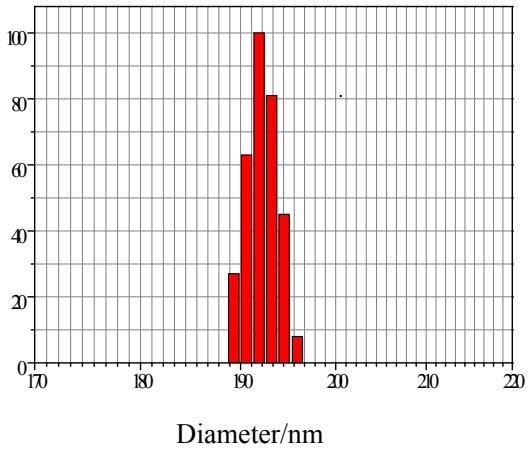

Figure S1. DLS of the NPAM-silica emulsion.

The size of the silica could be controlled by adding different amounts of surfactants. Here, we chose 0.2 CMC of the surfactant typically. The emulsion was stabilized by amino, and the silica was highly mono-dispersed with the size about 200 nm characterized by DLS. It was consistent with TEM and SEM results.

2. Temperature dependence of loss modulus of NPAM-silica/PU

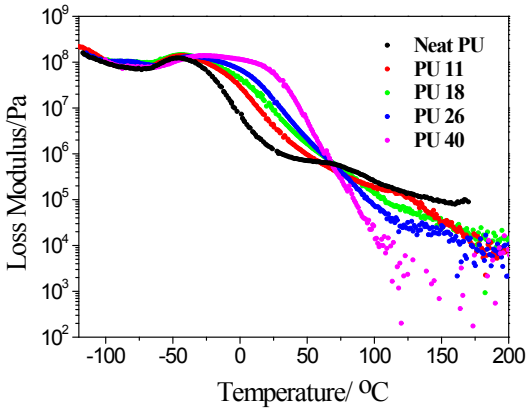

Figure S2. Loss moduli of NPAM-silica/PU composites.

Temperature dependence of loss modulus of NPAM-silica/PU was plotted as shown in Figure R2. It was obvious to see that the T_gs of soft segments of the

composites were all around -45 °C except an increase of T_g of PU40.

Table S1. Summarization of T_gs of both soft segments and hard segments

Sample	T_g (Soft segments) / from loss modulus - T curves*	T_g (hard segments)/ from tanδ – T curves*
Neat PU	-48 °C	-12 °C
PU11	-44 °C	7 °C
PU18	-43 °C	14 °C
PU26	-43 °C	22 °C
PU40	-34 °C	40 °C

*T_gs of the soft and hard segments of the NPAM-silica/PU nanocomposites were obtained from different methods. For T_gs of soft segments, they were more obvious in the loss moduli-T curves, while more obvious in the *tan δ*-T curves for T_gs of hard segments.