## Supporting Information

## Hydrogen Bond Regulating Miscibility of Graphene Oxide and Nonionic Water-Soluble Polymers

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Table S1. The miscibility rule of GO and PVA over the concentrated range.



Fig S1. (a) Chemical structure of GO and PVA. (b) XPS spectrum of GO.



**Fig S2.** Statistical results of the size of GO sheets with (orange) and without (blue) adding PVA. The mean size of the former is 39  $\mu$ m and the latter is 23  $\mu$ m. The sampling number is over 200 sheets.



**Fig S3.** Digital and OM images of aggregated GO-PVA mixture with low concentration at room temperature before (a) and after ultrasonication (b). Scale bars are 100 um.



**Fig S4.** OM images of inhomogeneous high concentration GO-PVA mixture (a) and homogeneous pure GO(b). Scale bars are 100 um.



Fig S5. Critical PVA content in GO under diifferent alholysis degree of 88% and 99%.



**Fig S6.** Digital images of GO-PVA mixture at different mass fraction and molecular weight and fixed alcoholysis degree of 88% (a) and 99% (b).



**Fig S7.** OM images of GO-PVA mixture at different mass fraction and molecular weight and fixed alcoholysis degree of 88% (a) and 99% (b). Scale bars are 100 um.



**Fig S8.** OM images of GO-PVA mixture at different mass fraction over the concentrated range at fixed PVA with molecular weight of 22 kDa and alcoholysis degree of 88%. Scale bars are 100 um.



Fig S9. SEM images of GO-PVA mixture at different temperature.



**Fig S10.** Digital (a) and OM (b) images of homogeneous GO-PVA (114.4 kDa and aloholysis degree of 88%) mixture after preheating. Scale bars are 100 um.



**Fig S11.** Digital and OM images of aggregated GO-PVA mixture by heating and stirring for 3h after cooling at 5 °C. Scale bars are 100 um.



Fig S12. The surface height profile image of P-GO-PVA (a) and R-GO-PVA (b).



**Fig S13.** Stress-strain curves of P-GO-PVA and R-GO-PVA with different GO content in PVA.



**Fig S14.** Digital photos and SEM images of transverse cross section of homogeneous GO-PVA-SiO<sub>2</sub> (a) and aggregated GO-PVA-SiO<sub>2</sub> (b).



**Fig S15.** Typical stress-strain curves (a) and mechanical properties (b) of homogeneous GO-PVA-SiO<sub>2</sub> and aggregated GO-PVA-SiO<sub>2</sub>.



**Fig S16.** Digital and optical microscope images of homogeneous GO-PVA -SiO<sub>2</sub> dispersions with different molecular weight of PVA.



**Fig S17.** Digital and optical microscope images of homogeneous GO-PVA -Gly dispersions with different molecular weight of PVA.



**Fig S18.** Digital and optical microscope images of GO-PEO, GO-HEC and GO-PVP dispersions before and after preheating at 85  $^{\circ}$ C.



**Fig S19.** Digital and optical microscope images of homogeneous GO-PVP/PEO/HPC -SiO<sub>2</sub>.



**Fig S20.** Digital and optical microscope images of homogeneous GO-PVP/PEO/HPC -Gly.

C <sub>GO</sub> (%)	C <sub>PVA</sub> (%)			
	0.1	0.5	1	5
0.001	$\checkmark$	$\checkmark$	$\checkmark$	$\checkmark$
0.01	×	$\checkmark$	$\checkmark$	$\checkmark$
0.1	×	×	×	$\checkmark$
0.5	×	×	×	×
1	×	×	×	×

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Notes: " $\sqrt{}$ " means miscible and " $\times$ " means aggregated