Electronic Supplementary Information (ESI)

High Lubricity and Electrical Responsiveness of Solvent-Free Ionic SiO₂ Nanofluids

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(S1) Structure characteristic of SiO₂-DC5700-NPES and SiO₂-PMPS-M2070 NFs by FTIR spectra

Figure S1a compared the FTIR spectra of initiator SiO₂ NPs, SiO₂-DC5700 salt and SiO₂-DC5700-NPES NFs. The broad peak at 3425 cm⁻¹ in the spectrum of SiO₂ NPs is assigned to the H-O-H stretching modes of the free or adsorbed water. The peak at 1105 cm⁻¹ can be attributed to the Si-O-Si anti-symmetric vibrations. Another two characteristic peaks at 812 and 473 cm⁻¹ correspond to the symmetric stretch and bending mode of the Si-O-Si, respectively¹. Three absorption peaks appearing at 2850 cm⁻¹, 2925 cm⁻¹ (-CH₂- stretching) and 1736 cm⁻¹ (C-N stretching) in the SiO₂-DC5700 are all exact proofs that DC5700 has grown from the initiator particles. Whereas, the peaks at 1646 cm⁻¹ and 1470 cm⁻¹ are assigned to the stretching vibration of the solvent-free SiO₂ NFs has been strengthened, which is ascribed to the stretching vibration of -CH₂- and -CH₃- from NEPS. The above described results corroborated that the SiO₂ initiator were grafted in succession with corona and canopy.

Figure S1b gives the FTIR spectra of SiO₂-PMPS salt and solvent-free SiO₂-PMPS-M2070 NFs. As for SiO₂-PMPS salt, the main absorption peaks at around 2948 and 1073 cm⁻¹ correspond to stretching vibrations of C-H and P=O, respectively, owing to grafting of PMPS onto the SiO₂ surface. Strong peaks at 2928 and 2860 cm⁻¹ are observed in the FTIR spectrum of SiO₂-PMPS-M2070 NFs, which are ascribed to the stretching vibrations of -CH₂- and -CH₃- from M2070. Additionally, the peak at 1057 cm⁻¹ is assigned to the C-O stretching vibration of M2070. These results give evidence of chemical bonding between SiO₂ NPs, PMPS and M2070.

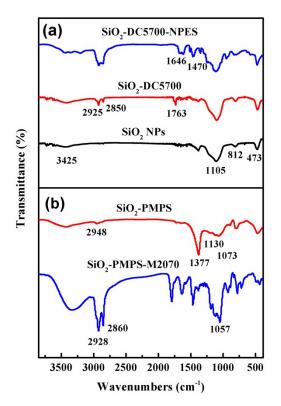


Figure S1. FTIR spectra of initiator SiO₂ NPs, SiO₂-DC5700 salt and SiO₂-DC5700-NPES NFs (a) and SiO₂-PMPS, SiO₂-PMPS-M2070 NFs (b).

(S2) TGA analysis of SiO₂-DC5700-NPES and SiO₂-PMPS-M2070 NFs

Figure S2 shows the TGA curves of the solvent-free ionic SiO_2 NFs, it is seen clearly that the solvent-free SiO_2 NFs exhibited different thermal stabilities owing to the distinct organic corona and canopy structures. At temperatures lower than 180 °C, S2

obvious mass loss was not noticed for both kinds of solvent-free NFs, indicating that the NFs endowed with good thermal stabilities and did not contain any residual solvent. However, SiO₂-DC5700-NPES NFs started to degrade at temperatures higher than 200 °C probably due to decomposition and oxidative degradation of lowmolecular-weight fragments in the canopy polymer. With respect to SiO₂-PMPS-M2070, no mass loss was noticed at temperatures lower than 300 °C, indicating its higher thermal stability than that of SiO₂-DC5700-NPES NFs. The rather high thermal stability SiO₂-PMPS-M2070 may allow potential applications under high temperature. Nonetheless, above 300 °C, both kinds of NFs started to degrade due to decomposition of the significant fraction of organic groups².

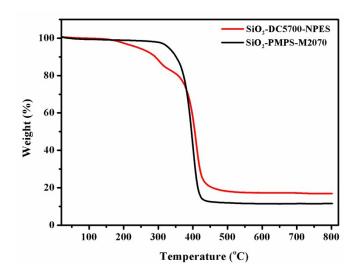


Figure S2. TGA curves of the solvent-free SiO₂ NFs

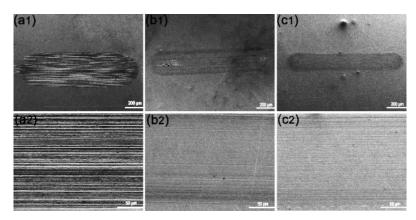


Figure S3. SEM micrographs of worn surfaces of the steel disks when lubricated with PEG (a1, a2), and PEG containing 5 wt% (b1, b2), 10 wt% (c1 and c2) solvent-free

SiO₂.DC5700-NPES NFs.

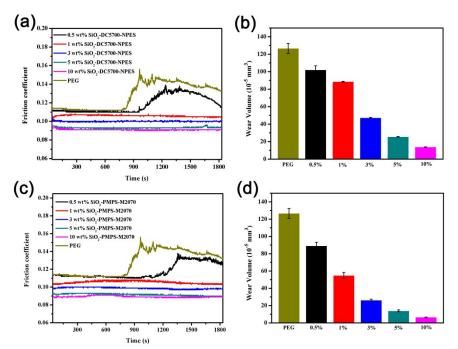


Figure S4. (a) Friction coefficients and (b) wear volumes of the steel disks lubricated with PEG and PEG containing SiO₂-DC5700-NPES NFs additive at 100 N; (c) friction coefficients and (d) wear volumes of the steel disks lubricated with PEG and PEG containing SiO₂-PMPS-M2070 NFs additive at 100 N.

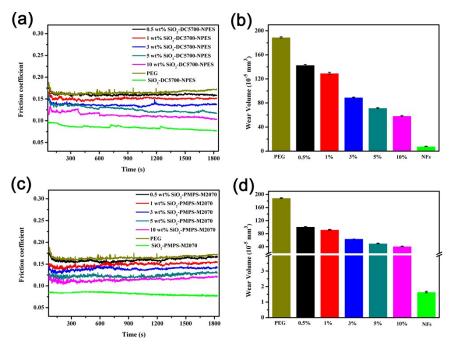


Figure S5. (a) Friction coefficients and (b) wear volumes of the steel disks lubricated with PEG and PEG containing SiO₂-DC5700-NPES NFs additive under 100 °C; (c) friction coefficients and (d) wear volumes of the steel disks lubricated with PEG and PEG containing SiO₂-PMPS-M2070 NFs additive under 100 °C.

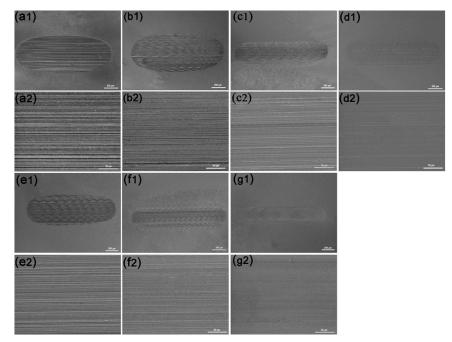


Figure S6. SEM micrographs of worn surfaces of the steel disks when lubricated with PEG (a1, a2); and PEG containing 1 wt% SiO₂-DC5700-NPES NFs (b1, b2); 5 wt% SiO₂-DC5700-NPES NFs (c1, c2); SiO₂-DC5700-NPES NFs (d1, d2); 1 wt% SiO₂-PMPS-M2070 NFs (e1, e2); 5 wt% SiO₂-PMPS-M2070 NFs (f1, f2) and neat SiO₂-PMPS-M2070 NFs (g1, g2) under 100 °C.

References

1 G. H. Du, Z. L. Liu, X. Xia, Q. Chu and S. M. Zhang, *J. Sol-Gel Sci. Technol.*, 2006, **39**, 285-291.

2 H. P. Bai, Y. P. Zheng, T. Y. Wang and N. K. Peng, *J. Mater. Chem. A*, 2016, **4**, 14392-14399.