Electronic Supplementary Information

Synergistic Effect of Organomodification and Isocyanate Grafting of Layered Double Hydroxide in Reinforcing Properties of Polyurethane Nanocomposites

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Fig. S1. FTIR spectra of (a) dry THF and (b) dried MDI-g-St-LDH/PU nanocomposite after swelling.

Fig. S2. X-Ray diffraction Patterns of pristine LDH, MDI-g-pristine LDH, IPDI-g-pristine LDH in the 2θ range of 10-80°.
**Fig. S3.** X-Ray diffraction patterns of (a) DS-LDH, MDI-g-DS-LDH, IPDI-g-DS-LDH and (c) St-LDH, MDI-g St-LDH and IPDI-g St-LDH in the $2\theta$ range of 10-80°.

**Fig. S4.** FTIR spectra of (a) MDI and (b) IPDI.
**Fig. S5.** FTIR spectra of physical mixtures of (a) DS-LDH with MDI (b) DS-LDH with IPDI, (c) St-LDH with MDI and (d) St-LDH with IPDI.

**Fig. S6.** FTIR spectra of (a) MDI-g-St-LDH and (b) its suspension in water after 72 h.
**Fig. S7.** $^1$H NMR spectra of (a) MDI-g-DS-LDH and (b) IPDI-g-DS-LDH.

**Fig. S8.** FTIR spectra of (a) prepolymer, prepolymer with (b) MDI-g-DS-LDH, (c) MDI-g-St-LDH, (d) IPDI-g-DS-LDH and (e) IPDI-g-St-LDH.
**Fig. S9.** Stress-strain plots of neat PU and its nanocomposite with 3 wt % DS-LDH and St-LDH.

**Grafting Calculation (wt %):**

The degree of grafting by isocyanate and modification by surfactant in LDHs and mass ratio of isocyanate, organomodifier and LDHs have been calculated from Wt. residues at 1000 °C in air which is shown in Table 5.

The weight residue of pristine LDH and their IPDI and MDI grafted LDHs are 53.68, 48.23 and 50.35 wt % i.e., MDI and IPDI grafted LDHs contain 89.84 (= 48.23/53.68) and 93.79 (=50.35/53.68) wt % LDH respectively. It is easy to estimate the respective degree of grafting of pristine LDH by MDI and IPDI are about 10.16 and 6.21 wt %.

Therefore, the mass ratio of IPDI-g-LDH is 50.35:6.21 i.e., 0.066:1 of IPDI:LDH and MDI-g-LDH contains 48.23:10.16 i.e, 0.113:1 of MDI:LDH.

DS-LDH contains (1- 41.46/53.68) = 22.77 wt % DS, 77.23 wt % LDH, giving a mass ratio of 0.294:1 :: DS:LDH.
MDI-g-DS-LDH contains (1 - 32.37/41.46) = 21.93 wt % MDI, 78.07 wt % DS-LDH (which is 77.23 wt % LDH and 22.77 wt % DS), giving 60.29 wt % LDH and 17.77 wt % DS, i.e. mass ratio of 0.363:0.294:1:: MDI:DS:LDH.

Similarly, for IPDI-g-DS-LDH contains (1 - 35.63/41.46) = 14.07 wt % IPDI, 85.93 wt % DS-LDH (which is 77.23 wt % LDH and 22.77 wt % DS), giving 66.36 wt % LDH and 19.56 wt % DS, i.e. mass ratio of 0.212:0.294:1:: IPDI:DS:LDH.

St-LDH contains (1-32.13/53.68) = 40.15 wt % St, 59.85 wt % LDH, giving a mass ratio of 0.670:1 :: St:LDH.

MDI-g-St-LDH contains (1 - 20.17/32.13) = 37.23 wt % MDI, 62.77 wt % St-LDH (which is 59.85 wt % LDH and 40.15 wt % St), giving 37.56 wt % LDH and 25.20 wt % St, i.e. mass ratio of 0.99:0.67:1:: MDI:St:LDH.

IPDI-g-St-LDH contains (1 - 24.65/32.13) = 23.28 wt % IPDI, 76.72 wt % St-LDH (which is 59.85 wt % LDH and 40.15 wt % St), giving 45.91 wt % LDH and 30.80 wt % St, i.e. mass ratio of 0.507:0.67:1:: IPDI:St:LDH.

Fig. S10. Thermogram of neat PU and its nanocomposites with 3 wt % DS-LDH and St-LDH.
Fig. S11. DSC scans of PU nanocomposites with 3 wt % DS-LDH and St-LDH.