Electronic Supplementary Information

Revealing the Three-Dimensional Filler Structure in Rubber Matrix Based on Fluorescein Modified Layered Double Hydroxides

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Fig. S1 ¹H NMR spectra of SBR 1502. The signal assignments are indicated.

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**Fig. S2** Fluorescent micrograph of the supernatant solution (SBR solution) derived from the dissolved 20 wt% SBR/F-NLDH compounds. The fluorescent intensity is weak and will not affect the observation of F-LDH particles in the rubber matrix.
Fig. S3 The critical strain ($\gamma_c$) as a function of LDH content ($\phi_r$) for (a) SBR/F-NLDH and (b) SBR/F-MLDH compounds.
**Fig. S4** Glass transition temperature of the F-NLDH filled compounds with various $\phi_{F}$. 
Fig. S5 (a) Reversing heat capacity of the rubber fraction in the F-MLDH filled compounds with various $\phi_r$. (b) Immobilized layer amount as a function of $\phi_r$. The insert shows the $\Delta C_p$ of compounds as a function of rubber fraction.
Fig. S6 Glass transition temperature of the F-MLDH filled compounds with various $\phi_f$. 

![Graph showing glass transition temperature vs LDH content](image-url)
Fig. S7 LSCM observation of the 10 wt% SBR/F-NLDH compounds before and after stretched to the strain of 100%. The histograms are the size distribution of LDH aggregates.
Fig. S8 LSCM observation of the 20 wt% SBR/F-NLDH compounds before and after stretched to the strain of 100%. The histograms are the size distribution of LDH aggregates.
Fig. S9 LSCM observation of the 10 wt% SBR/F-MLDH compounds before and after stretched to the strain of 100%. The histograms are the size distribution of LDH aggregates.
Fig. S10 LSCM observation of the 20 wt% SBR/F-MLDH compounds before and after stretched to the strain of 100%. The histograms are the size distribution of LDH aggregates.
Fig. S11 Schematic representation of the rubber chains before and after stretching. The chains are stretched and a strong slip on the filler surface is observed.