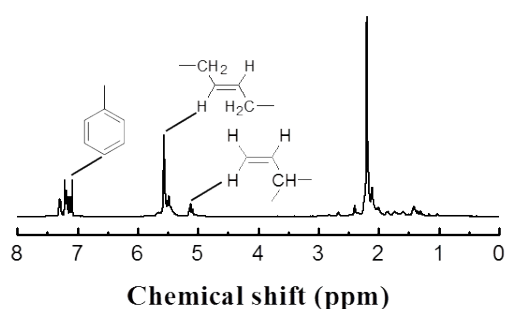


## Electronic Supplementary Information

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

Weiyang Lv, Miao Du\*, Jianliang Xiao, Yihu Song, Qiang Zheng

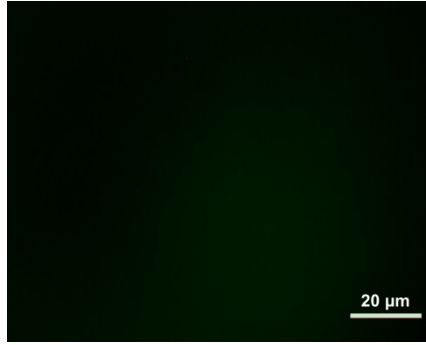
*MOE Key Laboratory of Macromolecular Synthesis and Functionalization, Department of Polymer Science and Engineering, Zhejiang University, Hangzhou, 310027, China*



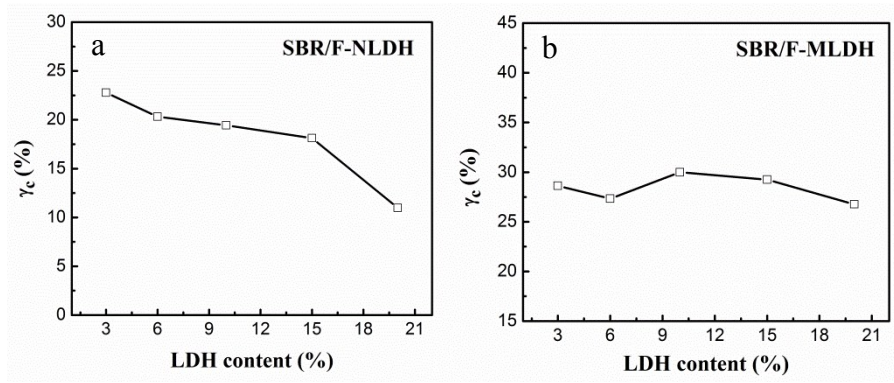
**Fig. S1** <sup>1</sup>H NMR spectra of SBR 1502. The signal assignments are indicated.

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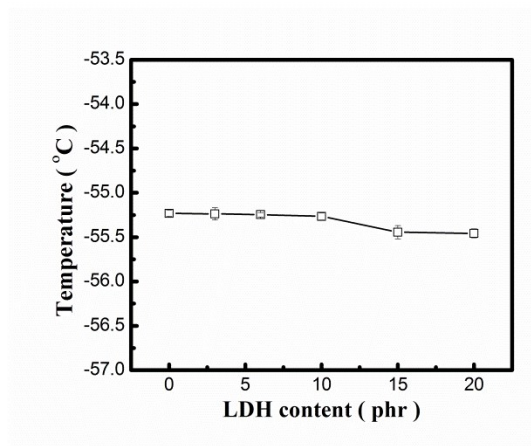
\*Corresponding author. E-mail: dumiao@zju.edu.cn, Tel.&Fax: 0086-571-87953075



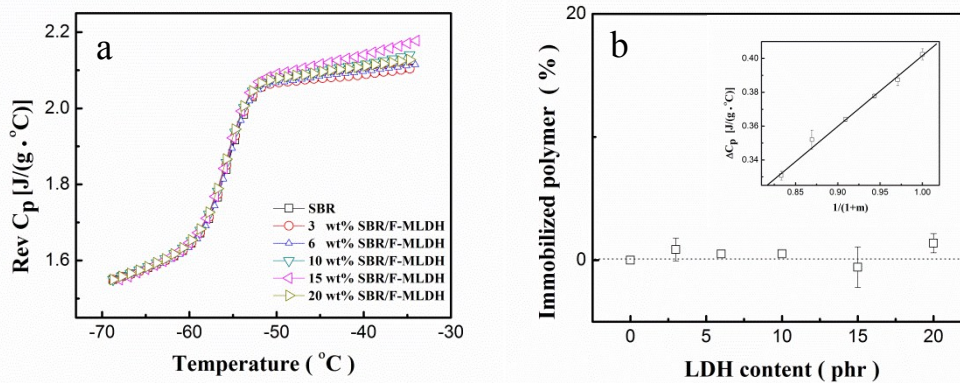
**Fig. S2** Fluorescent micrograph of the supernatant solution (SBR solution) derived from the dissolved 20 wt% SBR/F-NLDH compounds. The fluorescent intensity is weak and will not affect the observation of F-LDH particles in the rubber matrix.



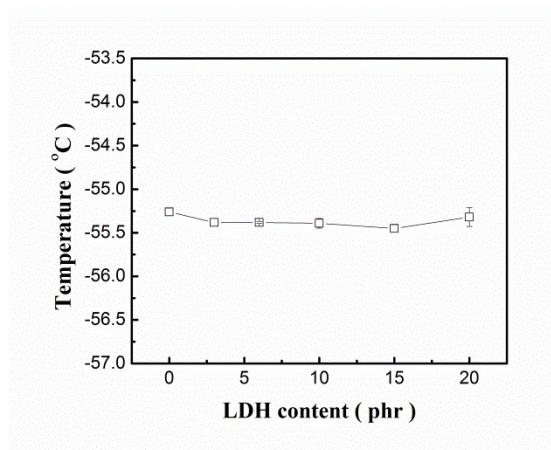
**Fig. S3** The critical strain ( $\gamma_c$ ) as a function of LDH content ( $\phi_F$ ) for (a) SBR/F-NLDH and (b) SBR/F-MLDH compounds.



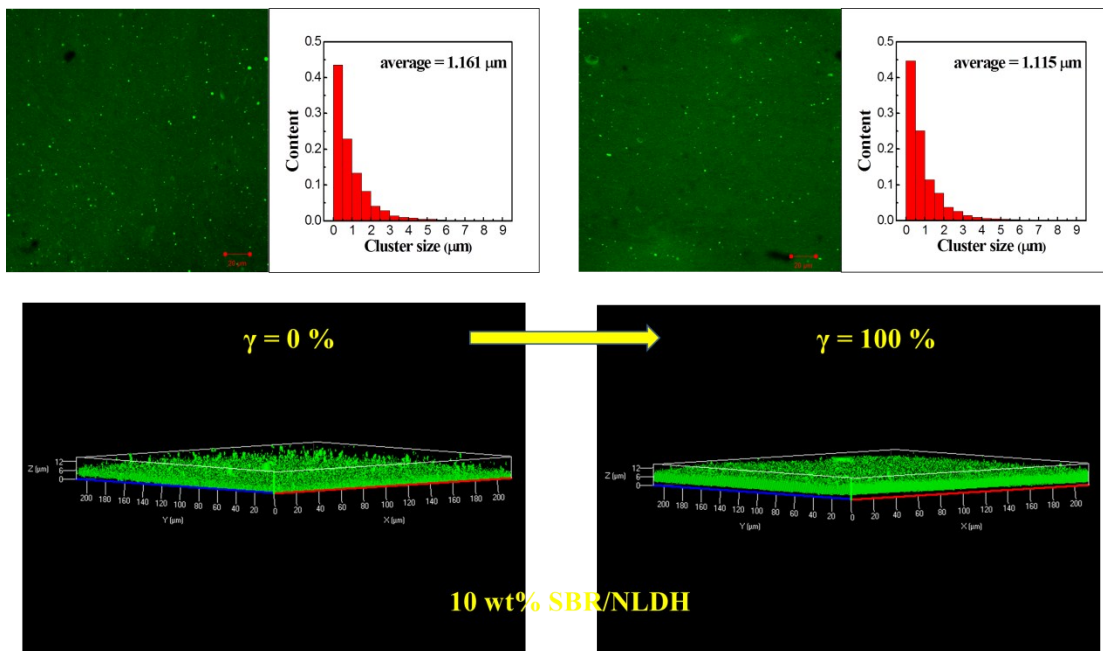
**Fig. S4** Glass transition temperature of the F-NLDH filled compounds with various  $\phi_F$ .



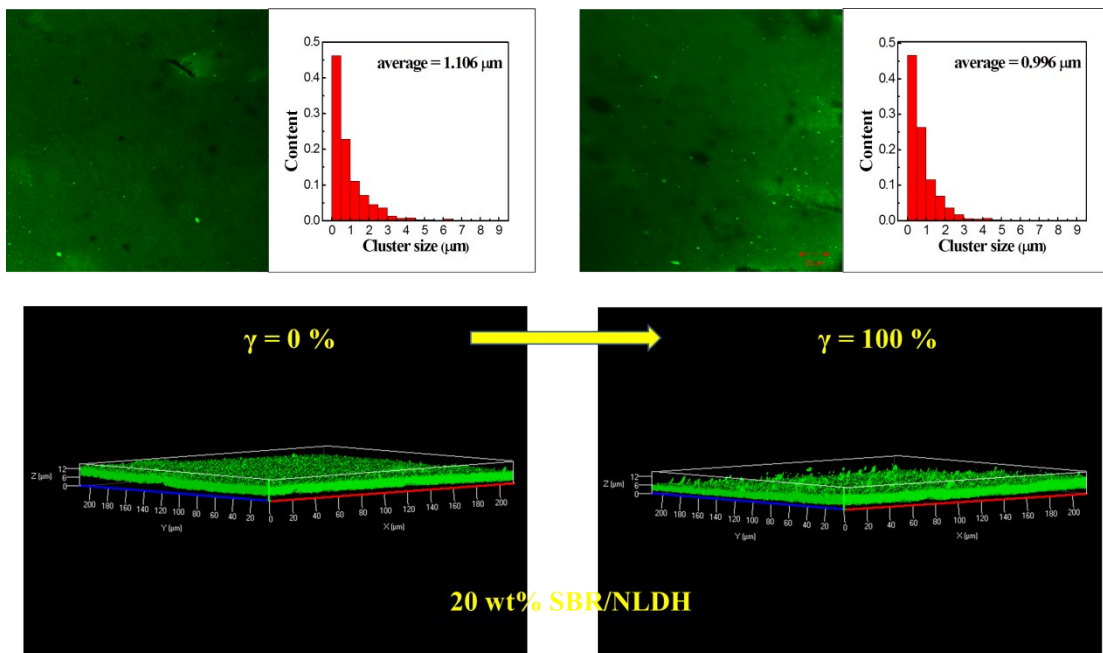
**Fig. S5** (a) Reversing heat capacity of the rubber fraction in the F-MLDH filled compounds with various  $\phi_F$ . (b) Immobilized layer amount as a function of  $\phi_F$ . The insert shows the  $\Delta C_p$  of compounds as a function of rubber fraction.



**Fig. S6** Glass transition temperature of the F-MLDH filled compounds with various  $\phi_F$ .

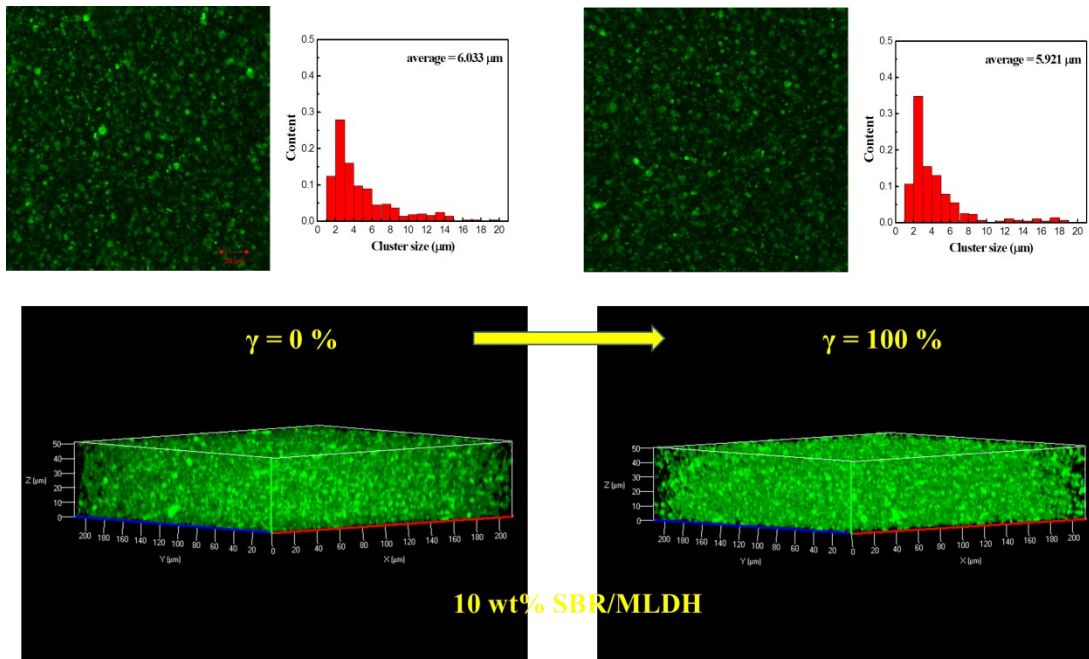


**Fig. S7** LSCM observation of the 10 wt% SBR/F-NLDH compounds before and after stretched to the strain of 100%. The histograms are the size distribution of LDH aggregates.

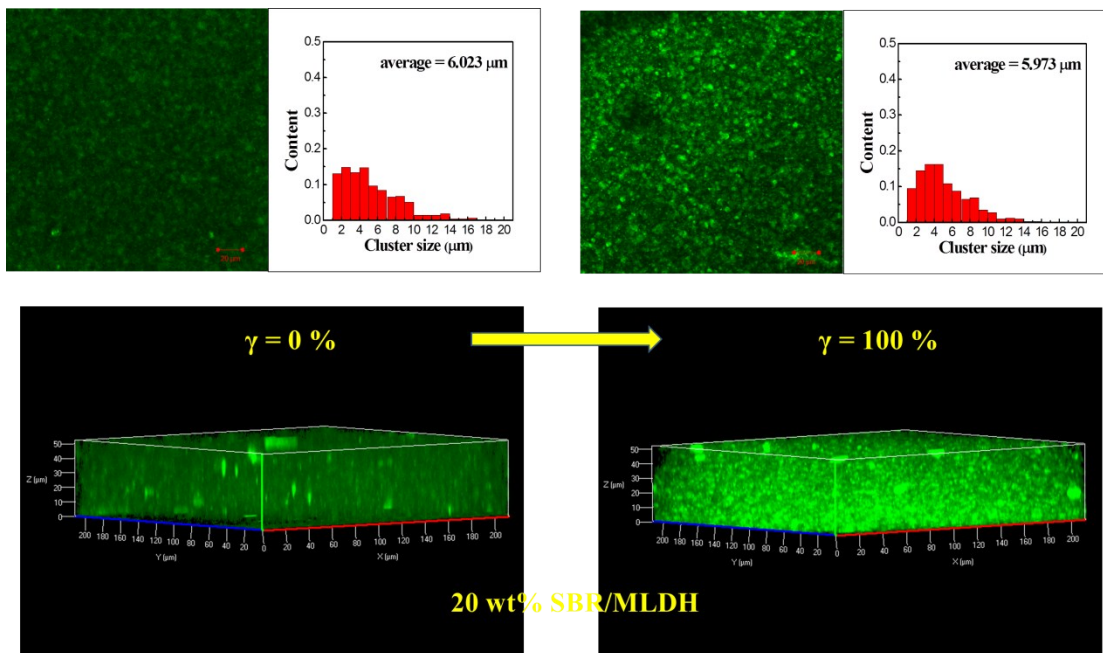


**Fig. S8** LSCM observation of the 20 wt% SBR/F-NLDH compounds before and after stretched to the strain of 100%. The histograms are the size distribution of LDH aggregates.

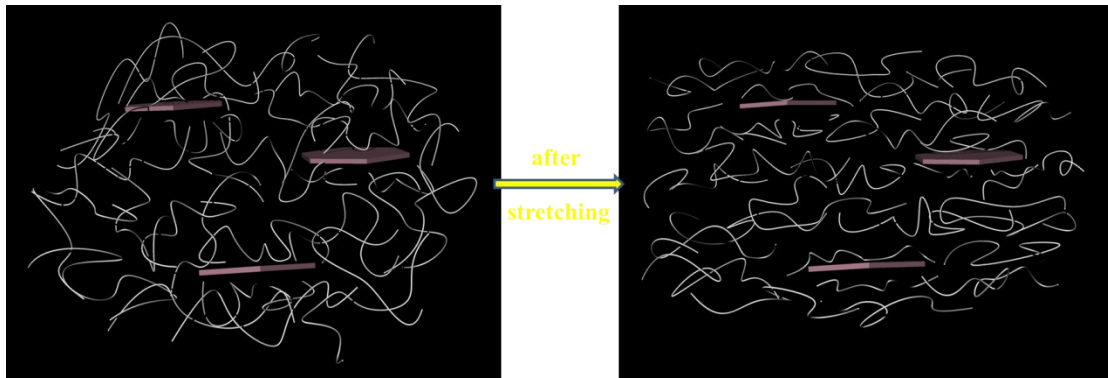




**Fig. S9** LSCM observation of the 10 wt% SBR/F-MLDH compounds before and after stretched to the strain of 100%. The histograms are the size distribution of LDH aggregates.



**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.