

Supplementary Materials

Highly Strong and Damage-Resistant Natural Rubber Membrane via Self-Assembly and Construction of Double Network

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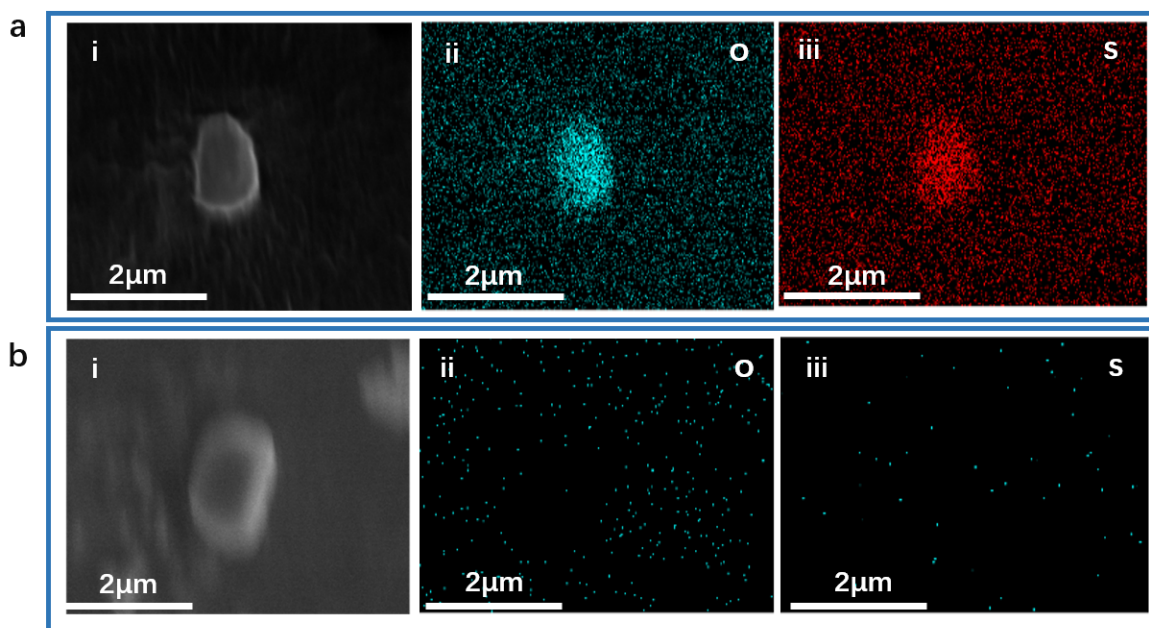


Figure S1. Element scan image of element distribution of (a) MSLS/NRL and (b) NRL latex particles.

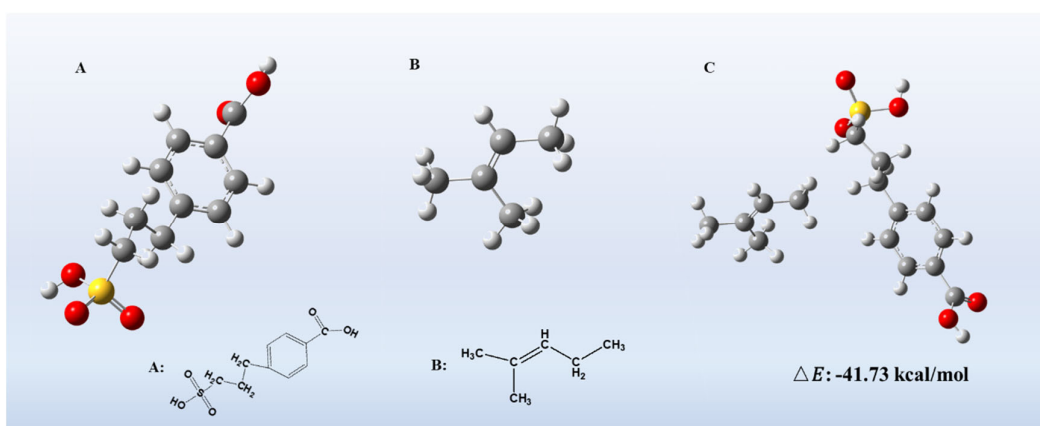


Figure S2. Density functional theory (DFT) calculation evaluating interaction energy between the MLSL and natural rubber.

To further study the core-shell formation mechanism between MLSL and rubber the interaction energy ($\Delta E_{\text{interaction}}$) was investigated by molecular dynamics simulation. DFT

calculation was conducted at ω B97X-D/6-31g** using G09 Software [1, 2]. Consequently, the interaction energy was calculated from the following equation [3]:

$$\Delta E_{interaction} = (E_{AB} - (E_A + E_B)) \quad (1)$$

Where E_{AB} and E_A (and E_B) are the total potential energies of final system and initial system, respectively.

Note: Structure-A and B were used to represent MLSL and natural rubber segment, respectively.

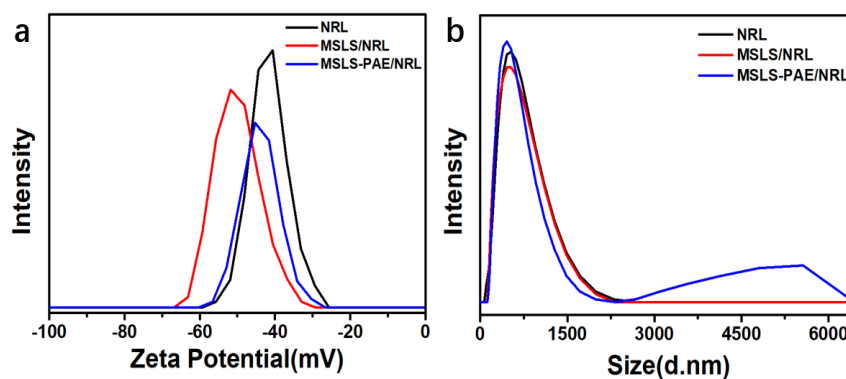


Figure S3. (a) Zeta potential and (b) particle size distribution of NRL, MSLS/NRL and MSLS-PAE/NRL.

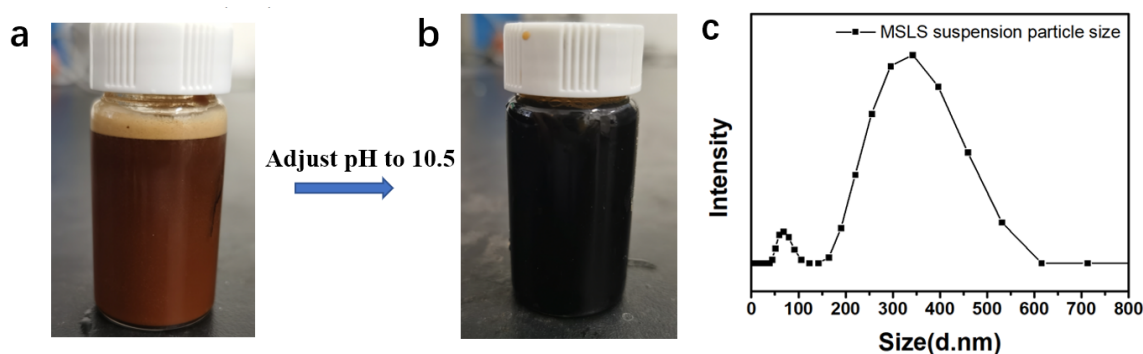


Figure S4. (a) During the oxidation, a picture of MSLS small brown particles, (b) a picture of MSLS suspension appear and the (c) particle size distribution of MSLS suspension.

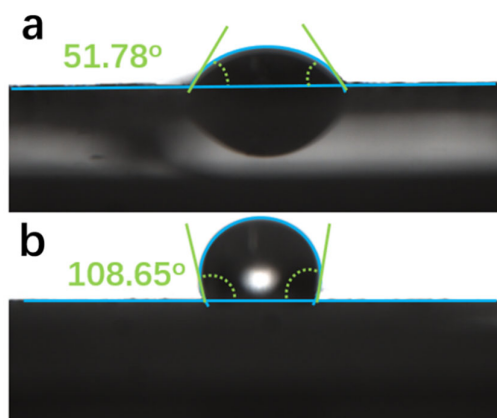


Figure S5. Water contact angle of (a) SLS and (g) MSLS.

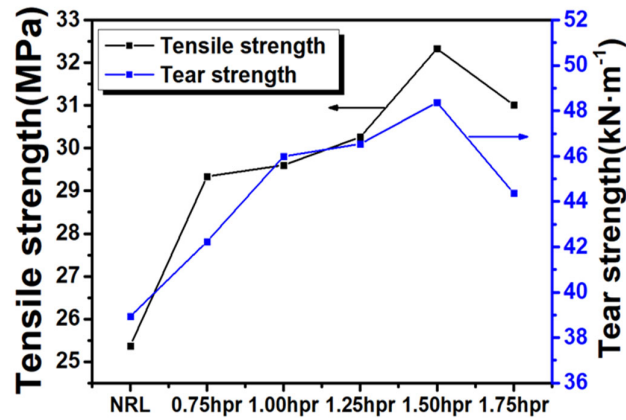


Figure S6. Tensile strength and tear strength of the composites with different MSLS content in natural rubber latex.

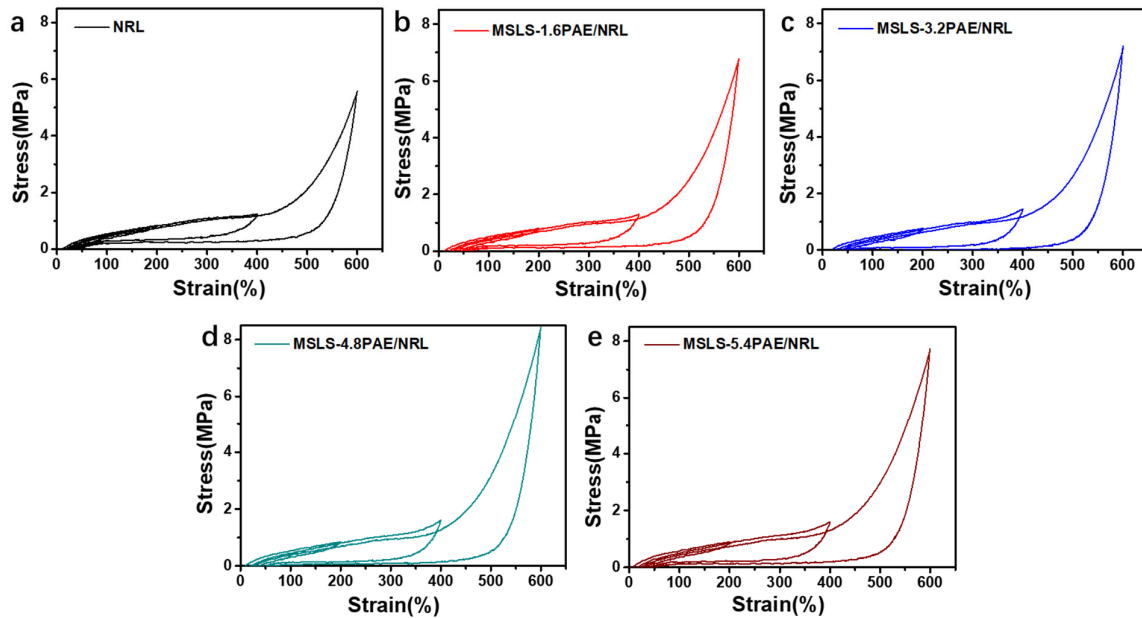


Figure S7. (a–e) the energy dissipation curves of rubber composites with different PAE contents.

Table S1. Zeta potential and particle size of NRL, MSLS/NRL and MSLS-PAE/NRL.

sample	Zeta Potential (mV)	Size (d. nm)
NRL	-46.37 ± 0.84	425.37 ± 8.70
MSLS/NRL	-52.90 ± 0.56	435.20 ± 17.49
MSLS-PAE/NRL	-48.57 ± 0.07	444.93 ± 5.34

Table S2. Estimated bond strength from reported references.

Bond type	Bond Energy(kJ/mol)	Ref.
C–C	370.8	[4]
C–O	344.5	[4]

C–N	338.9	[4]
S–S	267.1	[5]

Table S3. Statistics of mechanical properties of different materials.

Materials	Elongation at break (%)	Stress (MPa)	Elastic Modulus	Tear strength (kN·m ⁻¹)
NRL	771.50	25.37	3.29	41.39
MSLS-1.6PAE/NRL	761.50	32.57	4.28	48.48
MSLS-3.2PAE/NRL	809.13	34.69	4.29	55.23
MSLS-4.8PAE/NRL	884.38	37.27	4.21	61.08
MSLS-6.4PAE/NRL	789.13	33.45	4.24	57.12

References

1. Golder, M.R.; Wong, B.M.; Jasti, R. Photophysical and theoretical investigations of the cycloparaphenylene radical cation and its charge-resonance dimer. *Chem. Sci.* **2013**, *4*, 4285–4291.
2. Chai, J.D.; Head-Gordon, M. Systematic optimization of long-range corrected hybrid density functionals. *J. Chem. Phys.* **2008**, *128*, 084106.
3. Šponer, J.; Hobza, P.; Leszczynski, J. Chapter 3—Computational Approaches to the Studies of the Interactions of Nucleic Acid Bases. In *Theoretical and Computational Chemistry*; Leszczynski, J., Ed.; Elsevier: Amsterdam, The Netherlands, 1999; Volume 8, pp. 85–117.
4. Beyer, M.K. The mechanical strength of a covalent bond calculated by density functional theory. *J. Chem. Phys.* **2000**, *112*, 7307–7312, <https://doi.org/10.1063/1.481330>.
5. Stern, M.D.; Tobolsky, A.V. Stress-Time-Temperature Relations in Polysulfide Rubbers. *Rubber Chem. Technol.* **1946**, *19*, 1178–1192, <https://doi.org/10.5254/1.3543255>.