## Improvement of Filler Aggregation in Catechol-Functionalized Epoxidized Natural Rubber Composites

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Abstract : Natural rubber (NR) or cis-1,4-polyisoprene is a renewable polymer derived from Hevea brasiliensis plants, which is widely utilized in various applications, such as the tire industry. In terms of rubber processing, carbon black (CB) is commonly used as a reinforcing filler. However, filler aggregation of CB in rubber products is one of the important problems, which is related to the complicated mixing in rubber manufacturing and high energy loss. So, the mussel-inspired mechanism has been used to solve the problem of filler aggregation in rubber composites. This research aimed to improve the carbon black dispersion in epoxidized natural rubber (ENR) composites through aromatic interactions such as  $\pi$ - $\pi$  stacking and cation- $\pi$ interactions. Initially, the epoxidation process was used for the modification of NR to produce ENR. Then, the ENR was mixed with catechol as dopamine (D) and carbon black (CB), respectively. In this study, the aromatic interactions were obtained between the benzene rings in D molecules on ENR chains, and the surface of CB, which were observed in Fourier transform infrared spectroscopy and Raman spectroscopy. The results indicated that the mechanical properties were increased because of the effect of filler reinforcement and aromatic interactions within the ENR composites. Notably, this phenomenon was confirmed using the small/wide angle X-ray scattering (SAXS/WAXS), which was in good agreement with the rubber processing analyzer and transmission electron microscopy results that the  $\pi$ - $\pi$  stacking and cation- $\pi$  interactions enhanced the CB dispersion in the ENR composites. Therefore, these results showed the tensile strength, Young's modulus, and energy-saving properties reached up to 140%, 90%, and 50%, respectively. Finally, this research provides a novel approach based on a mussel-inspired material to solve the CB aggregation problem in rubber products, resulting in the achievement of ENR composites with superior properties.

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1