Degradation behavior during mixing of silica-reinforced Natural Rubber: Changes of the dynamic responses

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Abstract: High shearing forces and temperature are applied during mixing of silica filled natural rubber (NR) for tire tread applications, in order to achieve the best possible filler-rubber interactions and a sufficient silanization reaction of the silica. Both thermal and mechanical conditions in the mixing process can lead to polymer degradation. The present work investigates NR degradation during mixing via monitoring changes of its viscoelastic behavior. Silica-filled NR compounds prepared by using various dump temperatures were investigated taking pure NR and gum or unfilled NR compounds as references. Chain scission and chain recombination as two competitive reactions affect the molecular weight and chain architecture. Chain scissions most likely contribute more to the viscous response whereas chain recombination and interactions contribute to the elastic component of viscoelasticity. Increasing viscous responses of masticated pure NR and gum compounds are observed with higher dump temperature as measured by Mooney stress relaxation rates, changes of storage and loss moduli, resp. tan δ with frequency, and large amplitude oscillatory shear (LAOS). Chain scission causes a lower molecular weight, but a broader molecular weight distribution and more branching. For silica-filled NR compounds, the elastic response rises at high dump temperatures above 150°C due to crosslinking and/or branching. The long-chain branching index (LCB) rises with increasing dump temperature. Furthermore, deterioration of tensile properties of the corresponding vulcanizates are observed which is attributed to degradation and chain modifications.

Keywords: natural rubber; degradation; silica-reinforcement; visco-elastic properties; tires