Elucidating degradation phenomena during mixing of silica-reinforced Natural Rubber compounds through changes of their dynamic responses

Ammarin Kraibut[1,2], Kannika Sahakaro[1], Sitisaiyidah Saiwari[1], Wilma K. Dierkes[2], Jacques W.M. Noordermeer[2]

[1] Department of Rubber Technology and Polymer Science, Faculty of Science and Technology, Prince of Songkla University, Pattani, Thailand

[2] Department of Sustainable Elastomer Systems, Faculty of Engineering Technology, University of Twente, Enschede, The Netherlands

Corresponding authors: Kannika Sahakaro; e-mail: kannika.sah@psu.ac.th and Jacques W.M. Noordermeer; e-mail: j.w.m.noordermeer@utwente.nl

Abstract: Mixing of silica-reinforced natural rubber (NR) compounds for tire applications requires appropriate shear forces to achieve the best possible filler-rubber interactions and optimization of the silanization reaction of the silica. The mixing process involves both thermal and mechanical input that can lead to polymer degradation. The present work focusses on the degradation of NR during mixing through changes of its viscoelastic behavior via dynamic testing. Silica-filled NR compounds subjected to different dump temperatures were tested taking pure NR and gum or unfilled NR compounds as references. Two major competitive reactions, i.e. chain scission and crosslinking or chain recombination affect the molecular weight and chain architecture. Shorter molecules by chain scission contribute more to the viscous response, while crosslinks and interactions contribute to the elasticity component. As analyzed by Mooney stress relaxation rates, changes of storage and loss moduli, resp. tan δ with frequency, large amplitude oscillatory shear (LAOS) measurements, the viscous responses of masticated pure NR and gum compounds gradually increase with rising dump temperature. The chain scission causes a lower molecular weight, a broader molecular weight distribution and more branching. For the silica-filled NR compounds, the elastic response rises at high dump temperatures above 150°C due to crosslinking and/or excessive branching. The long-chain branching index (LCB) is increased with increasing dump temperature. The degradation and chain modifications result in a reduction of tensile properties of the corresponding vulcanizates.

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