

# How Silane Coupling Agents Influence the Dynamic Performance of SBR/Silica Tire Tread Compounds

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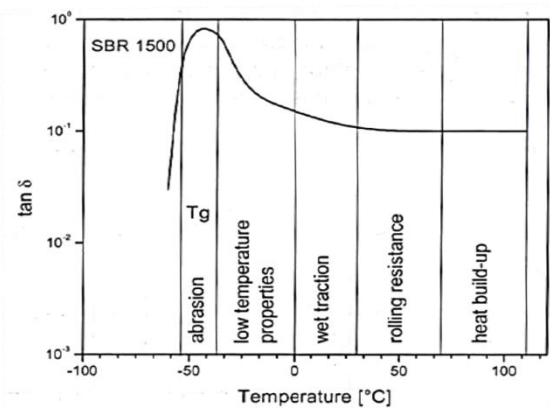
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## 1. Introduction

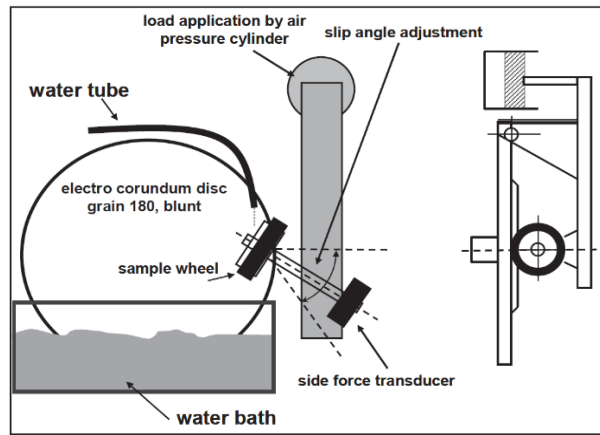
Rubber compounds used for tire tread applications are filler-reinforced composite materials of which the dynamic properties can be adjusted over a relatively broad range by modification of the polymer-filler interaction. The replacement of carbon black as reinforcing filler two decades ago by silica-silane systems allowed to reduce the low-frequency hysteresis of the rubber, which resulted in reduction of the Rolling Resistance (RR) when applied in tire treads. Apart from the selection of the proper elastomers and silica's for this technology, the choice of the coupling agents to chemically link the polar silica to the apolar elastomers is of paramount importance. Next to their reactivities towards the silica-particles and to the elastomer molecules in order to obtain sufficient coupling, the intricate chemical structure of the coupling agents once coupled does still influence the RR and the Wet Skid Resistance (WSR) or Wet Traction of the treads. Coupling agents commonly applied are variants of silane types. Selective changes in the structures of the silane coupling agents, e.g. de number of alkoxy-groups attached to the Si-atom reactive towards silanol-groups on the surface of the silica filler, the length of the aliphatic linker between the Si-atom and the reactive group towards the elastomer, or the bond strength between the coupling agent and the polymer, lead to changes in the microstructure of the silica-elastomer interface. All these changes determine the dynamic properties, as indicative for the RR as well as WSR and Abrasion Resistance of a tire tread. The paper primarily covers the RR and WSR, making use of dynamic mechanical analysis: laboratory scale indications for RR and WSR by means of the dynamic loss angle as a function of temperature, and of a practical laboratory assessment to specifically obtain an indication of WSR by means of a Laboratory Abrasion Tester 100 (LAT100).

## 2. Procedures and Results

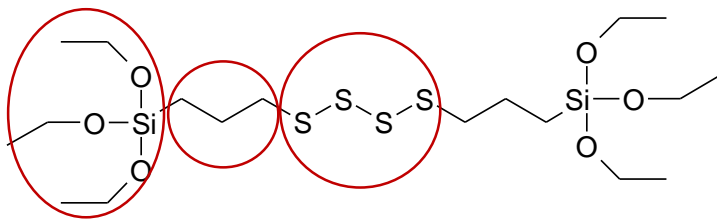
Figure 1 presents the commonly employed indications for RR and WSR based on the dynamic mechanical loss function  $\tan \delta = G''/G'$ , where  $G''$  resp.  $G'$  are the loss resp. storage moduli of a tire compound [1]. The validity of this approach to predict the WSR is subject to debate [2], and therefore use was made of the LAT 100 method for that prediction, a step closer to the actual skidding of the tire on the road. A picture and the basic principle of this equipment is illustrated in Figure 2.



**Fig. 1** Commonly accepted correlation of dynamic loss angle to primary tire performance functions.

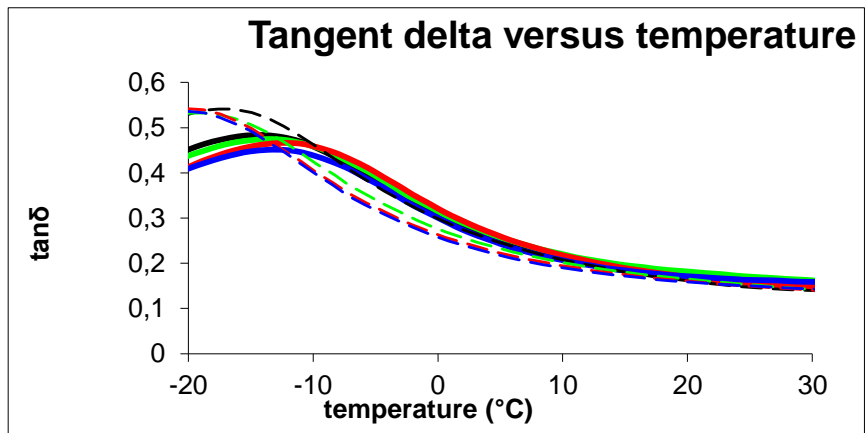


**Fig. 2** Illustration and basic principle of LAT 100 to simulate WSR as a function of temperature.



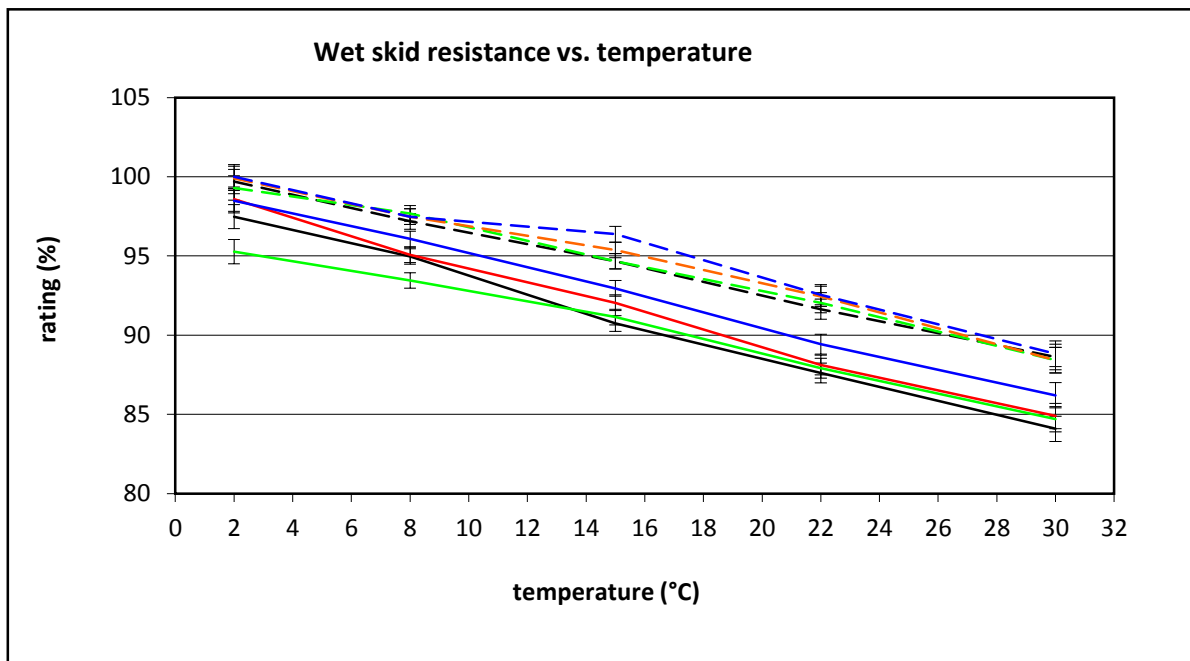
**Fig. 3** Basic structure of TESPT coupling agent

The types of coupling agents investigated were variations on the principle coupling agent bis-(tri-ethoxy silyl propyl)-tetrasulfide (TESPT), as depicted in Figure 3. Variations as indicated by red circles included: (1) No sulfur atoms in the structure: bis-(tri-ethoxy silyl)-hexane (TESH), which can only couple to the silica-surface; (2) bis-(tri-ethoxy silyl decyl)-tetrasulfide (TESDeT), which has a longer aliphatic linker which may enhance the segmental flexibility of the interface between silica and elastomer; (3) bis-(di-methyl mono-ethoxy silyl propyl)-tetrasulfide (DMESPT), which has only one ethoxy-group per Si-atom to react with silica, evolving reduced amounts of ethanol during the reaction with silica compared to the other two. The compound formulations were based on a commonly applied recipe for silica-reinforced passenger car tire treads: a synthetic S-SBR/BR blend with 80 phr of easy dispersion silica and 7 phr TESPT [3] and the others at equimolar quantities to be comparable with TESPT.

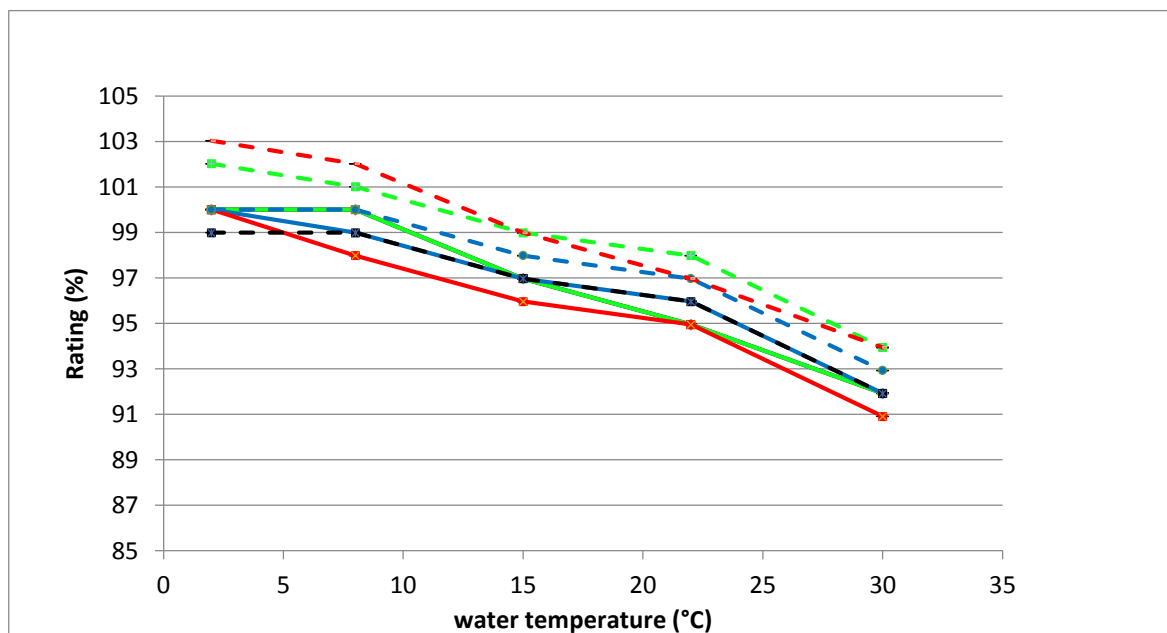


**Fig. 4**  $\tan\delta$  vs. temperature for various concentrations of TESH (solid lines) vs. TESPT (dashed lines).

Figure 4 presents an example of the dynamic mechanical loss function variation with temperature for equimolar amounts of TESH and TESPT: the first for 5.3, 7.3, 9.3 and 11.3 phr; the latter for 7, 9.5, 12 and 14.4 phr. Based on the assumed predicting power of these measurements for WSR it would be anticipated, that the higher  $\tan\delta$  around 0 °C would indicate an improved WSR for TESH relative to TESPT. However, the LAT 100 experiments presented in relative rating in Figure 5 indicate the opposite: TESPT performs better in WSR than TESH in the LAT100. Similar comparisons will be presented for TESDeT.



**Fig. 5** Wet skid resistance vs. temperature for 4 loadings of TESH (solid lines) vs. 4 loadings of TESPT (dashed lines) as recorded with LAT 100.



**Fig. 6** WSR vs. temperature for DMESPT (dashed lines) vs. TESPT (solid lines).

The results for DMESPT vs. TESPT differ from the other two: DMESPT combines a better WSR as measured in the LAT 100, see Figure 6, with an improved lower RR [3] for most of the silane loadings, as shown in Figure 7. Tentatively this may be interpreted in terms of a more organized surface coverage of the silica by the DMESPT than by TESPT, while only one ethoxy-group per Si-atom is provided for reaction with silica, rather than 3, and because less ethanol is evolved during the silica-silane coupling which tends to remain partly on the silica surface, and so provides a more stable interface.

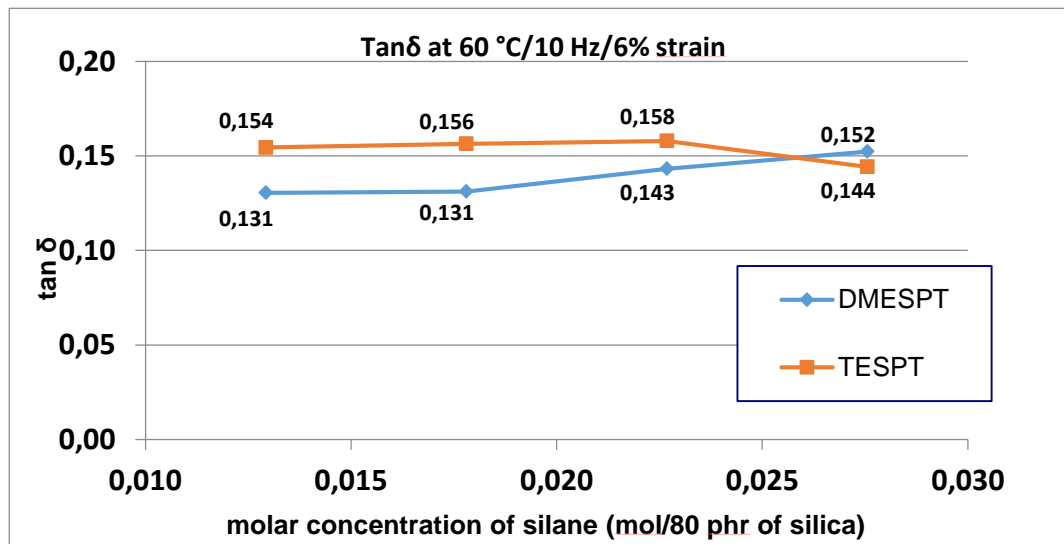


Fig. 7  $\tan \delta$  at 60 °C as a measure for RR for various loadings of DMESPT and TESPT.

### 3. Conclusions

- Wet Skid Resistance and Rolling Resistance deteriorate when the coupling agent TESH is employed, without sulfur in the structure and so unable to couple to the elastomer phase;
- Increased linker length relative to the propyl-linker as within TESDeT reduces/improves the indications for Rolling Resistance (not shown), but has no effect on Wet Skid Resistance;
- Using a coupling agent with just one ethoxy-functionality per Si-atom: TMESPT, vs. 3 in TESPT, reduces/improves the indications for Rolling Resistance, as well as indicates an improved Wet Skid Resistance in applied in tire tread compounds.

### 4. Acknowledgements

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