

Effects of Silane on Properties of Silica Filled Natural Rubber Compounds

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The Mooney viscosity and cure properties of some conventional accelerator/sulphur compounds of natural rubber containing, respectively, 10, 30 or 60 parts per hundred rubber by weight (p.p.h.r.) precipitated amorphous white silica were measured. The compounds also contained no silane, and different amounts of bis[3-triethoxysilylpropyl-] tetrasulphane bifunctional organosilane. The viscosities of the compounds with 60 p.p.h.r. and 30 p.p.h.r. silica reduced substantially when 7 p.p.h.r. and 1.5 p.p.h.r. silane were added, respectively. The viscosity of the mix with 10 p.p.h.r. silica decreased only slightly as the dosage of silane was raised to 5 p.p.h.r. The shortest optimum cure times, t_{95} , for the mixes with 60 p.p.h.r. and 30 p.p.h.r. silica, were recorded when 7 p.p.h.r. and 4 p.p.h.r. silane were added respectively, whereas, the cure time of the compound with 10 p.p.h.r. silica was little affected by the addition of up to 5 p.p.h.r. silane. The cure rate of the mixes with 60 p.p.h.r. and 30 p.p.h.r. silica, calculated from the cure rate index, accelerated significantly, when 7 p.p.h.r. and 4 p.p.h.r. silane were added, respectively. Interestingly, the cure rate of the mix with 10 p.p.h.r. silica, slowed down greatly as a result of introducing up to 5 p.p.h.r. silane.

For many years, colloidal carbon blacks have been utilised to reinforce properties of rubber products such as car tyres, hoses and antivibration mounts. When added to the formulation, these fillers help to improve physical and mechanical properties; for example, tear resistance, hardness and abrasion resistance, of rubber compounds. Synthetic silicas such as precipitated silicas, have been replacing carbon blacks, to some extent, in many industrial applications, offering similar benefits. Precipitated silicas are prepared by a variety of techniques such as precipitation from water-soluble silicates¹, and also from chemical reaction between a sodium silicate

solution and sulphuric acid². The surfaces of silicas possess siloxane and silanols chemical functional groups³, and the silanol or hydroxyl groups are acidic⁴. Moreover, because the surface of the filler is polar and hydrophilic, there is a strong tendency to adsorb moisture^{5,6}. The amount of water adsorbed on the surface of the filler controls the ionisation of the hydroxyl groups⁷, resulting also in detrimental effects on the cure attributes of rubber compounds. These unique properties create numerous problems when silicas are used in rubber products. For instance, when a large amount of silica is added, the viscosity increases significantly, causing detrimental

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effects on the processibility of the compounds, and also excessive wear and tear of the processing equipment. Studies⁸ have shown that when large quantities of precipitated silica were added to natural rubber compounds, increases of compound viscosities through the incorporation of the filler could be attributed primarily to the strong silica-silica interactions. In addition, acidic silicas interact with basic accelerators, causing detrimental effects such as unacceptably long cure times and slow cure rates³, and also loss of crosslink yield⁹ in sulphur cure systems. Wagner¹ reported large increases in the scorch and optimum cure times of some conventional accelerator/sulphur compounds of natural rubber, when the loading of precipitated silica was raised progressively. For example, the cure time, t_{90} at 150°C, lengthened from 9 min to 41 min, when the amount of silica was increased from 20 p.p.h.r. to 60 p.p.h.r., respectively. He also noted that the viscosity of the compounds rose from 16 to 91 Mooney units, when this amount of silica was added¹.

To promote a wider use of silicas in industrial rubber products, it is essential to remedy these processing problems. Bifunctional organosilanes are designed for use in sulphur cure systems³. These materials help reduce viscosity by weakening or diminishing silica-silica interactions³, and improve cure characteristics by preventing acidic silica from interfering with the reaction mechanism of sulphur cure systems³. Preliminary studies by the author and co-workers^{10,11} showed that adding silane to silica filled sulphur/accelerator cure system-based natural rubber compounds, helped to reduce viscosity, and shorten scorch and cure times of the mixes, considerably.

The aim of this extended study was to examine how a progressively increasing amount of a bifunctional organosilane in the

formulation, influenced the Mooney viscosity and cure properties of some conventional accelerator/sulphur compounds of natural rubber containing different amounts of precipitated silica. We aimed mainly at optimising the silane in these compounds, and ultimately maximising the benefits of this additive to the viscosity and cure characteristics of these mixes. This, in turn, will help to improve the design properties of such compounds, and keep the costs of materials and processing under control.

EXPERIMENTAL

Materials and Mixing

Fifteen conventional accelerator/sulphur compounds of natural rubber, with a sulphur to accelerator ratio of about 4.17, were tested (*Table 1*). These compounds contained, respectively, 10, 30 or 60 p.p.h.r. precipitated amorphous white silica-type Ultrasil VN3[®], and also no silane, and different amounts of bis[3-triethoxysilylpropyl-)tetrasulphane (TESPT) bifunctional organosilane (*Table 1*). This silane is known as Si69 coupling agent, and is suitable for use in sulphur cure systems³. The silica and silane were supplied by Degussa AG, Germany. The silica was stored at 18°C and an ambient relative humidity of 76% for eight weeks prior to use. It has been suggested¹² that the most efficient method of using the silane is by pre-mixing with the filler before addition to the raw elastomer. Following this advice, the silica was sprayed with the silane in a glass flask at ambient temperature (~23°C) before adding to the raw elastomer. The rubber used was a Standard Malaysian Natural Rubber Grade L. In addition to the elastomer, silica and silane, some other ingredients were also added. These are listed in *Table 1*.

TABLE 1. FORMULATION FOR THE COMPOUNDS

Formulation (p.p.h.r.)	Compound number														
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
Natural rubber (SMR L)	100	100	100	100	100	100	100	100	100	100	100	100	100	100	100
Stearic acid	2	2	2	2	2	2	2	2	2	2	2	2	2	2	2
Santoflex 13 [®] (antidegradant)	1	1	1	1	1	1	1	1	1	1	1	1	1	1	1
Silane (TESPT)	0	1	2	3	5	0	1.5	2.5	4	6	0	3	5	7	10
Silica (Ultrasil VN3) [®]	10	10	10	10	10	30	30	30	30	30	60	60	60	60	60
Santocure [®] (CBS)	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6	0.6
Sulphur	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5	2.5
Zinc oxide	5	5	5	5	5	5	5	5	5	5	5	5	5	5	5

Definitions:

Silane (TESPT): Bis (triethoxysilylpropyl) tetrasulfane

Santocure[®] (CBS): N-cyclohexyl-2-benzothiazolesulphenamideSantoflex 13[®]: N- (1,4-demethylbutyl)-N'-phenyl-ppd**Mixing**

Mixing was carried out in a Francis Shaw K1 Intermixer Mark 4 with intermeshing counter-rotating-type rotors. In these experiments, the rotors and the mixing chamber were maintained at 40°C with a water cooling system. Chronos Richardson SOW 1000 software was utilised for controlling the mixing condition and storing data. The volume of the mixing chamber was 5.5 litres, and a fill factor of 0.5 was considered for preparing these compounds. For each compound, batch temperature was plotted against time on a linear-linear axis, and the ram position was also shown on the same graph (*Figure 1*). Full details of the mixing procedure are given in *Tables 2, 3 and 4*.

Test-Procedure

Characteristics of the silica. The moisture content of the silica was determined by weighing ~ 4.94 grams of the material, and placing it in a vacuum oven at 110°C under a pressure of 0.04 bar equivalent to 96% vacuum for 6 h to dry. The sample was subsequently removed from the oven, and placed in a desiccator with fresh silica gel for 10 min to cool down. After this time elapsed, the sample was recovered from the desiccator and immediately weighed on an electronic balance. Finally, the weight loss was calculated as a percentage of the initial weight of the sample.

Silica pH was measured at 20°C using a WPA CD 720 pH meter, after it was calibrated

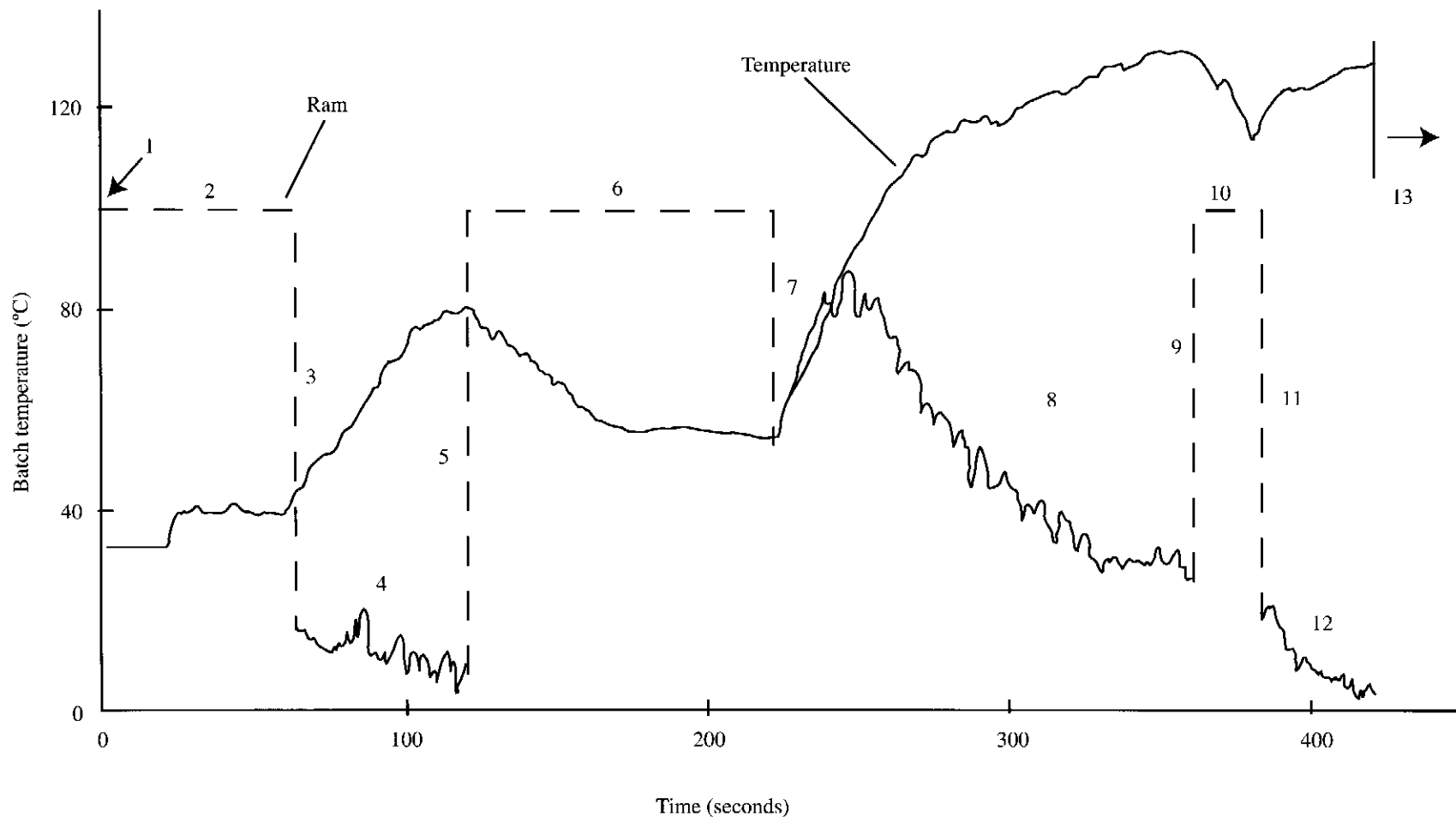


Figure 1. Typical record of batch temperature as a function of time for the compounds (—) and ram position (---). Data for the compound with 60 p.p.h.r. silica and 7 p.p.h.r. silane. Ram raised (1), raw elastomer, Santoflex® and stearic acid added (2), ram lowered (3), mixing (4), ram raised (5), silane treated silica added (6), ram lowered (7), mixing (8), ram raised (9), zinc oxide, Santocure® and sulphur added (10), ram lowered (11), mixing (12), batch discharged (13).

TABLE 2 MIXING CONDITIONS FOR THE COMPOUNDS WITH 10 PPHR SILICA

Formulation (pphr)	1	2	3	4	5
	Stage one mixing				
Time ram was raised (sec)	~31	~39	~37	~41	60
Ingredients added	Raw elastomer, Santoflex 13 [®] and stearic acid				
Ram lowered					
Compound temperature (°C) (when mixing started)	~57	~57	~58	~57	~58
Mixing time (sec)	~58	~59	~58	~58	60
Compound temperature (°C) (when mixing ended)	~69	~87	70	~81.5	~71
	Stage two mixing				
Time ram was raised (sec)	45	~35	~39	~37	~44
Ingredients added	Silane-treated silica				
Ram lowered					
Compound temperature (°C) (when mixing started)	~61	80	~64	~69	~64
Mixing time (sec)	~139	~137	~137	~137	140
Compound temperature (°C) (when mixing ended)	~124	~125	123.5	~125	120
	Stage three mixing				
Time ram was raised (sec)	~25	89	~35	~31	20
Ingredients added	Zinc oxide, Santocure [®] (CBS) and sulphur				
Ram lowered					
Compound temperature (°C) (when mixing started)	~106	~108	~101	~109	~109.5
Mixing time (sec)	~39	37.5	40	40	38
Compound temperature (°C) (when mixing ended)	~121	~124	119	~122	~118
Ram pressure 3 bar					
Rotor speed 45 rpm					

with pH 4 and 7 buffers. Five grams of silica was dispersed in 100 ml of de-ionised water, and stirred for one min. The pH of the

dispersion was further monitored for 5 min until it stabilised, and finally a reading was taken.

TABLE 3. MIXING CONDITIONS FOR THE COMPOUNDS WITH 30 P.P.H.R. SILICA

Formulation (p.p.h.r.)	6	7	Compound number 8	9	10 ^a
Stage one mixing					
Time ram was raised (sec)	~69	~26	~31	~16	—
Ingredients added:	Raw elastomer, Santoflex 13 [®] and stearic acid				
Ram lowered					
Compound temperature (°C) (when mixing started)	~61	42	~59	~55	—
Mixing time (sec)	~61	59	59	~59	—
Compound temperature (°C) (when mixing ended)	~81	75	~69	~57	—
Stage two mixing					
Time ram was raised (sec)	56.5	39	~29	~58	—
Ingredients added:	Silane-treated silica				
Ram lowered					
Compound temperature (°C) (when mixing started)	~79	~57	60	~57	—
Mixing time (sec)	140	~138.5	139.5	~138.5	—
Compound temperature (°C) (when mixing ended)	~131	~127	~127	~125	—
Stage three mixing					
Time ram was raised (sec)	~40.5	~19	~21	~32	—
Ingredients added:	Zinc oxide, Santocure [®] (CBS) and sulphur				
Ram lowered					
Compound temperature (°C) (when mixing started)	~111	~109	~108	~110	—
Mixing time (sec)	~38	~39	~38	37.5	—
Compound temperature (°C) (when mixing ended)	~127	123.5	~125	~122	—
Ram pressure: 3 bar					
Rotor speed: 45 r.p.m.					

^aNo mixing data are available for this compound

Properties of the compounds. The Mooney viscosity, [ML(1+4)], of the compounds was measured at 100°C, in a single-speed rotational

Mooney viscometer¹³. The scorch time, t_{s2} (time to two units rise in torque above minimum torque) (Figure 2), and optimum

TABLE 4. MIXING CONDITIONS FOR THE COMPOUNDS WITH 60 P.P.H.R. SILICA

Formulation (p.p.h.r.)	Compound number				
	11	12	13	14	15
Stage one mixing					
Time ram was raised (sec)	30	~31	~42	~61	~19
Ingredients added:	Raw elastomer, Santoflex 13 [®] and stearic acid				
Ram lowered					
Compound temperature (°C) (when mixing started)	~50	~48	40	40	~53
Mixing time (sec)	60	60	60	60	60
Compound temperature (°C) (when mixing ended)	~62	~61	~69	80	~66
Stage two mixing					
Time ram was raised (sec)	~79	~45	~129	~103	99
Ingredients added:	Silane-treated silica				
Ram lowered					
Compound temperature (°C) (when mixing started)	~60	~53	~51	~55	~61
Mixing time (sec)	140	—	140	140	140
Compound temperature (°C) (when mixing ended)	140	—	~135	~132	~134
Stage three mixing					
Time ram was raised (sec)	~47	—	25	~19	~26
Ingredients added:	Zinc oxide, Santocure [®] (CBS) and sulphur				
Ram lowered					
Compound temperature (°C) (when mixing started)	~110	—	~113	114	~125
Mixing time (sec)	40	—	40	40	40
Compound temperature (°C) (when mixing ended)	~148	—	~131	~130	~134
Ram pressure: 3 bar					
Rotor speed: 45 r.p.m.					

cure time, t_{95} (time to 95% of maximum torque development) (Figure 2), were calculated from the following equation¹⁴:

$$T_{\text{new}} = 0.95 (T_{\text{max}} - T_{\text{min}}) + T_{\text{min}} \quad \dots 1$$

where T_{max} is maximum torque, and T_{new} is a new torque reading corresponding to 95% cure in the rubber determined from the cure traces (i.e. Figure 2), generated at $140^{\circ}\text{C} \pm 2^{\circ}\text{C}$ by Oscillating Disc Rheometer Curemeter at an

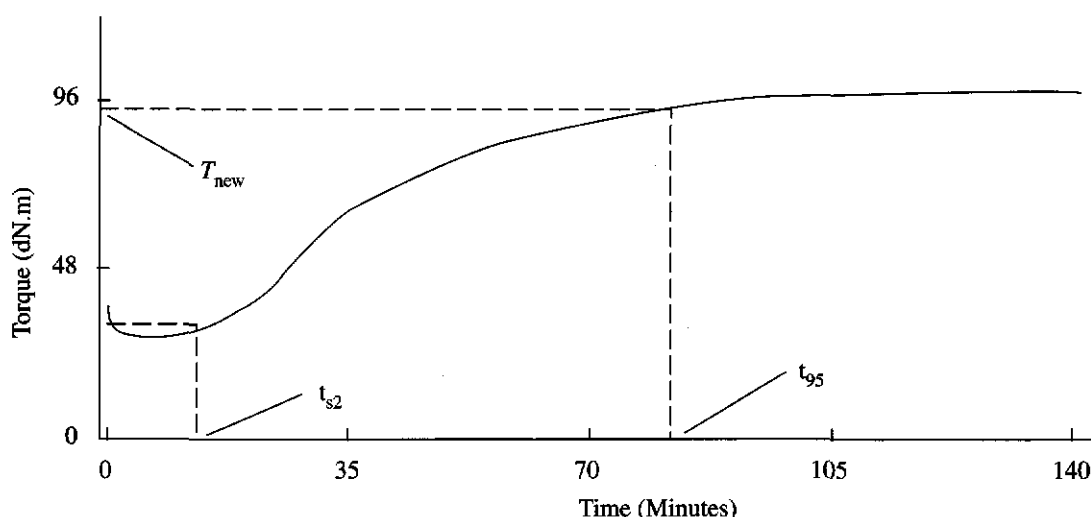


Figure 2. Typical torque versus time trace by ODR at 140°C. Data for the compound with 60 p.p.h.r. silica and 7 p.p.h.r. silane showing scorch time, t_{s2} , and optimum cure time, t_{95} .

angular displacement of $\pm 3^\circ$ and a frequency of 1.7 Hz¹⁴. The cure rate index, which is proportional to the average slope of the cure rate in the steep region of the cure curve, was also calculated using the following expression¹⁵:

$$100/(t_{95} - t_{s2}) \quad \dots 2$$

RESULTS AND DISCUSSION

The Mooney viscosities, scorch times, optimum cure times, and cure rate indices of the compounds are listed in Table 5. These results are also presented in a graphical form in Figures 3 to 6.

Figure 3 shows Mooney viscosity, [ML(1+4)], as a function of silane content. The viscosity of the compound with 60 p.p.h.r. silica decreases almost by a half, from about 234 units to 121 units, when 7 p.p.h.r. silane is introduced,

and remains at this level thereafter, when the loading of silane increases to 10 p.p.h.r. Similarly, the viscosity of the mix with 30 p.p.h.r. silica drops to 100 units from 129, after only 1.5 p.p.h.r. silane is added. It is worth noting that further increases in the amount of this additive to 6 p.p.h.r. appears to have little effect on this measurement, which is only slightly changed to about 91 units. Interestingly, the addition and progressive increases in the dosage of silane to 2 p.p.h.r., has almost no effect on the viscosity of the compound with 10 p.p.h.r. silica, which remains at about 80 units. It is also evident from Figure 3 that when the amount of silane is raised by an extra 3 p.p.h.r. to 5 p.p.h.r., the viscosity of this compound decreases only by 10 units to about 70 units. Wolff⁹ likewise reported marked reduction; from 100 to about 50 Mooney units, in the viscosity of some natural rubber compounds containing 40 p.p.h.r. precipitated silica, when he added 2.5 p.p.h.r. TESPT silane. In more recent studies¹⁷, Ismail observed

TABLE 5. RESULTS FROM THE MOONEY VISCOSITY MEASUREMENTS AT 100°C, AND THE ODR TESTS CARRIED OUT AT 140°C

	Compound number														
	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15
Mooney viscosity ML (1+4) at 100°C	80	82	80	77	71	129	100	98	95	91	234	175	146	121	119
	ODR results														
Minimum torque (dNm)	22.5	22	20	20.5	17	33	28	28	27	25	64	43	34	30	31
Maximum torque (dNm)	55	57	57	60	55	56	53	64	69	73	81	92	86	99	102
Scorch time, t_{82} (min)	26.5	23	21	18	18.5	9	18	15	15	14	34	7.5	10.2	12.3	14.4
Optimum cure time, t_{95} (min)	52	49	48	47	58	168	130	90	74	71	178	115	105	82	95
Cure rate index (min^{-1})	3.92	3.85	3.70	3.45	2.53	0.63	0.89	1.33	1.71	75	0.69	0.93	1.05	1.43	1.24

noticeable reduction in the Mooney viscosity of some rubber mixes loaded with 30 p.p.h.r. precipitated silica, when 3 p.p.h.r. mercaptosilane was incorporated into the formulation. This behaviour is somewhat similar to what we discovered with the mixes containing 30 p.p.h.r. and 60 p.p.h.r. filler loading (*Figure 3*).

Figures 4 and 5 show scorch time, t_{82} , and optimum cure time, t_{95} , as a function of silane loading, respectively. The scorch time of the mix with 60 p.p.h.r. silica, shortens from 34 min to about 7.5 min; this time may be too long for the industrial processing of these compounds and could be made shorter by adding polyethylene glycol or diethylene glycol to the formulation, when 3 p.p.h.r. silane is introduced, but it slowly lengthens to about 15 min as the loading of silane is raised progressively to 10 p.p.h.r. In contrast, adding

1.5 p.p.h.r. silane raises the scorch time of the mix with 30 p.p.h.r. silica, from 9 min to 18 min. However, as the amount of silane increases to 6 p.p.h.r., the scorch time shortens by 4 min to about 14 min. Evidently, the compound with 10 p.p.h.r. silica, requires up to 3 p.p.h.r. silane to reduce its scorch time from about 27 min to 18 min; the shortest scorch time recorded for this mix (*Figure 4*). Further increases in the quantity of silane to 5 p.p.h.r., appears to have little or no effect on the scorch time of this compound, which remains at about 18 min.

The optimum cure time, t_{95} , of the compounds are likewise influenced by the addition and progressive increases in the amount of silane (*Figure 5*). The cure time of the mix with 60 p.p.h.r. silica, decreases from 178 min to 82 min, when 7 p.p.h.r. silane is

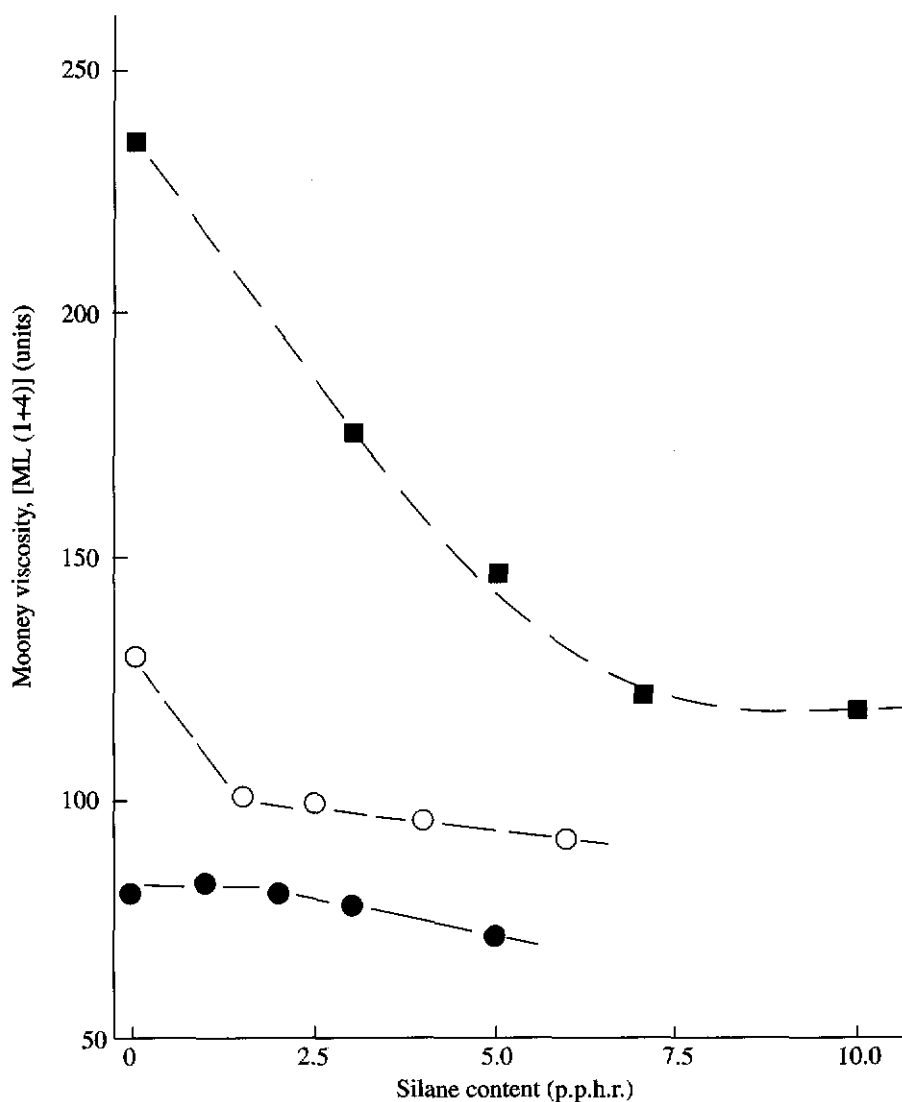


Figure 3. Mooney viscosity, [ML(1+4)] at 100°C, as a function of silane content. 60 p.p.h.r. silica (■); 30 p.p.h.r. (○); 10 p.p.h.r. silica (●).

added, but shows a modest rise to 95 min, after the dosage of this additive is boosted by an extra 3 p.p.h.r. to 10 p.p.h.r. It is interesting to note that a similar trend is also recorded

for the mix with 30 p.p.h.r. silica, with the cure time shortening from 168 min to 74 min after 4 p.p.h.r. silane is mixed. Further increases in the loading of silane to 6 p.p.h.r.,

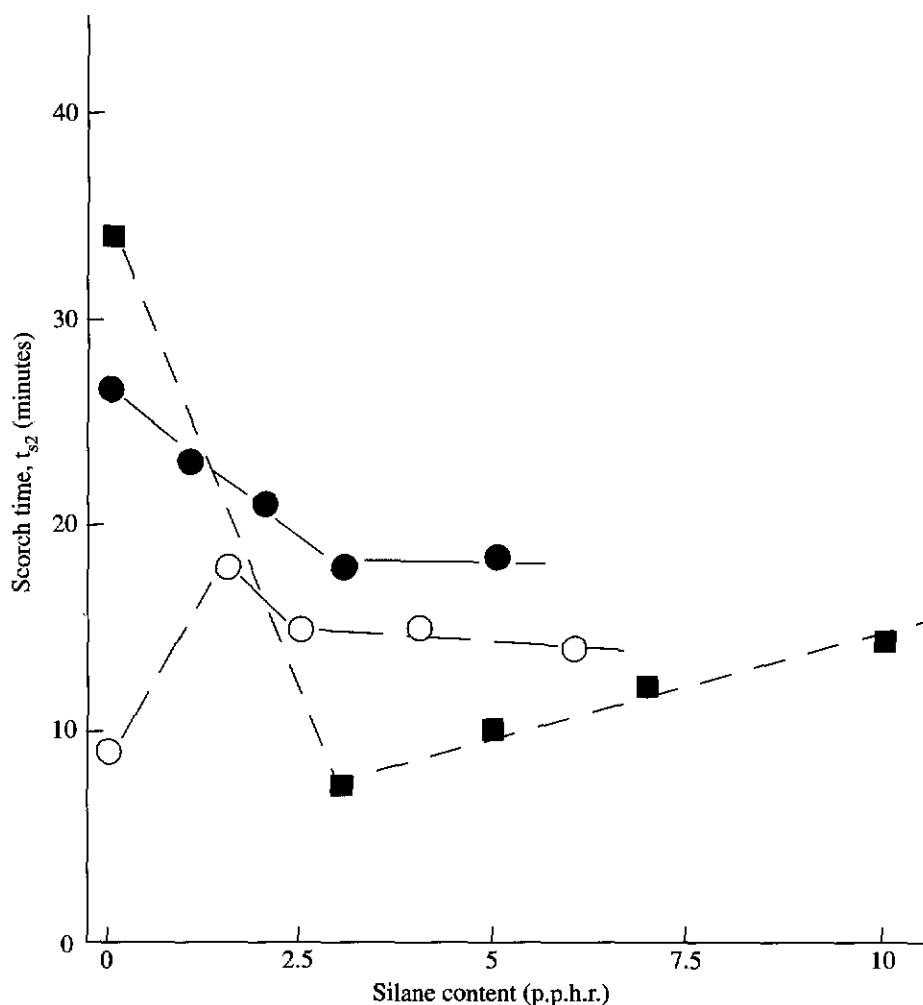


Figure 4. Scorch time, t_{s2} , as a function of silane content. 60 p.p.h.r. silica (■); 30 p.p.h.r. (○); 10 p.p.h.r. silica (●).

seems to offer little benefit to the cure time of the mix which stays at about 71 min. There is also an indication in Figure 5 that adding up to 3 p.p.h.r. silane reduces the cure time of the compound with 10 p.p.h.r. silica only slightly, from 52 min to 47 min. Notably, further increases of silane to 5 p.p.h.r. in this

compound, appears to have a detrimental effect on its cure time, which lengthens to 58 min, a rise of about 11 min (Figure 5).

It has been known³ for some time that silanes influence scorch and cure times to various degrees dependent on the accelerator

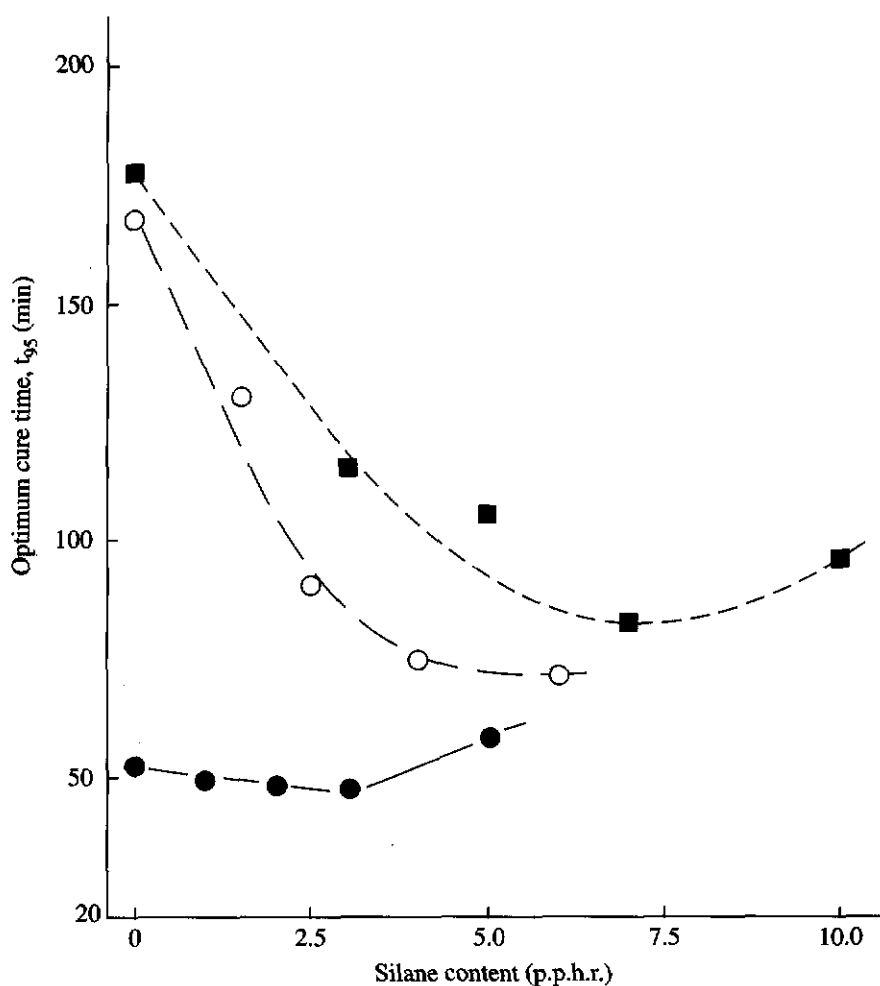


Figure 5. Optimum cure time, t_{95} , as a function of silane content. 60 p.p.h.r. silica (■); 30 p.p.h.r. silica (○); 10 p.p.h.r. silica (●).

system and elastomer used. Studies by Ranuey and co-workers¹⁶ have shown that adding 1 p.p.h.r. of mercaptosilane to a conventional sulphur/accelerator system-based silica-NR formulation; with a sulphur to accelerator ratio of about 2.3 and also containing 50 p.p.h.r. precipitated silica, reduced the scorch and cure

times from 30 min to 4 min and 30 min to 10 min, respectively. This trend is identical to some of our results for the mixes with 60 p.p.h.r. silica, where the scorch and cure times likewise shorten from 34 min to 7.5 min and 178 min to 115 min, respectively, when 3 p.p.h.r. silane is introduced (Table 5). In a

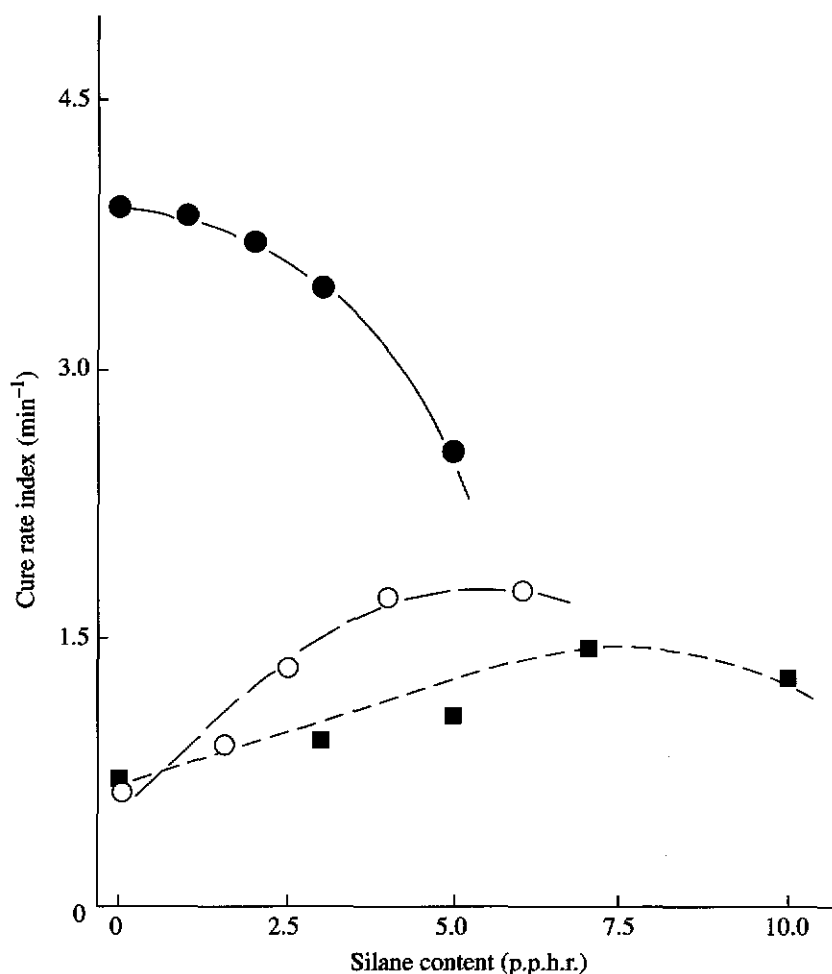


Figure 6. Cure rate index as a function of silane content. 60 p.p.h.r. silica (■); 30 p.p.h.r. silica (○); 10 p.p.h.r. silica (●).

similar study, Ismail¹⁷ discovered that the scorch and optimum cure times of some sulphur cure system-based natural rubber compounds containing 30 p.p.h.r. silica, decreased gradually and noticeably as the amount of silane increased progressively from 0 p.p.h.r. to 3 p.p.h.r. These findings are

in line with what we have learnt in this investigation.

The cure rate index which is a measure of the rate of cure in the rubber (Figure 6), increases from 0.69 min⁻¹ to 1.43 min⁻¹ for the mix with 60 p.p.h.r. silica after 7 p.p.h.r.

silane is added, suggesting that the rate of curing reaction in the rubber has accelerated significantly. However, this rate is little changed as the loading of silane is raised by an extra 3 p.p.h.r. to 10 p.p.h.r. A similar behaviour is also observed for the mix with 30 p.p.h.r. silica after 4 p.p.h.r. silane is introduced, though the rate of cure accelerates even faster in this compound, with the cure rate index increasing from 0.63 min^{-1} to 1.71 min^{-1} , but remaining at this level thereafter, when the loading of silane is raised further to 6 p.p.h.r. In contrast, when silane is added to the mix with 10 p.p.h.r. silica and increased progressively to 5 p.p.h.r., it retards the rate of cure noticeably, with the cure rate index decreasing from 3.92 min^{-1} to about 2.53 min^{-1} (Figure 6). It is worth noting that the rate of curing reaction in the mixes with 30 p.p.h.r. and 60 p.p.h.r. silica and no silane, are almost identical with cure rate indices of 0.63 min^{-1} and 0.69 min^{-1} , respectively (Figure 6).

As mentioned earlier on⁸, increases in compound viscosity when filler is added, are attributed mainly to strong silica-silica interaction, resulting from polar surface functional groups such as siloxane and silanols or hydroxyl groups^{3,4}. The subject silica possessed a moisture content of about 5.7% by weight, which corresponded to a surface hydroxyl concentration⁵ somewhere between 9.2 and 15.8 per 100 Å². The polarity of the filler surfaces resulting from the presence of these functional groups, has produced strong silica-silica interaction⁸, and hence increased the viscosities of the mixes with silica and no silane (Figure 3). The acidity of the filler⁴; the subject silica had a pH of 5.9, and the physically adsorbed moisture on the surfaces of silica; ~5.7% by weight, have retarded the rate of cure and prolonged the cure times of the compounds with silica and no silane significantly, as expected³. This effect on

properties is more detrimental for the mixes containing 30 p.p.h.r. and 60 p.p.h.r. silica (Figures 3–6).

It is abundantly clear from this study that adding TESPT silane to these compounds helps to remedy the processing problems discussed above, mainly by blocking off the acidic surface hydroxyl groups¹² and weakening filler-filler interaction. Moreover, adding silane has prevented these groups from interfering with the reaction mechanism of sulphur-cure system³ in these compounds. It has also emerged that to maximise benefits from TESPT silane, it is essential to use the appropriate amount of this additive in the formulation. This is essentially decided by the loading of silica in the rubber.

CONCLUSIONS

This study has showed that when up to 10 p.p.h.r. TESPT silane was added to conventional sulphur/ accelerator compounds of natural rubber, with a sulphur to accelerator ratio of 4.17 and also containing, respectively, 10, 30 or 60 p.p.h.r. precipitated amorphous white silica-type Ultrasil VN3[®], the viscosity and cure properties were profoundly affected, depending on the amount of silica present in the formulation.

The following was also evident:

- Mooney viscosities of the compounds containing 30 p.p.h.r. and 60 p.p.h.r. silica, decreased substantially, when 1.5 p.p.h.r. and 7 p.p.h.r. silane, respectively were added. The addition and progressive increase of silane to 5 p.p.h.r., offered little practical benefit to the viscosity of the mix with 10 p.p.h.r. silica.

- Scorch times of the compounds with 10 p.p.h.r. and 60 p.p.h.r. silica reduced to their minimum values after 3 p.p.h.r. silane was introduced. The time of mix with 30 p.p.h.r. silica to scorch off increased at first when 1.5 p.p.h.r. silane was added, and then reduced gradually and noticeably as the loading of silane was raised further to 6 p.p.h.r.
- Optimum cure times of the compounds containing 30 p.p.h.r. and 60 p.p.h.r. silica reduced to their lowest values, when 4 p.p.h.r. and 7 p.p.h.r. silane, respectively, were mixed. Notably, little advantage was gained by adding up to 5 p.p.h.r. silane to the mix with 10 p.p.h.r. silica.
- Rate of cure of the mixes containing 30 p.p.h.r. and 60 p.p.h.r. silica accelerated to its maximum level after 4 p.p.h.r. and 7 p.p.h.r. silane, respectively, were introduced. Surprisingly, the addition and progressive increase of silane to 5 p.p.h.r. appeared to have a detrimental effect on the cure rate of the mix with 10 p.p.h.r. silica.

In summary, major benefits can be gained in optimising silane in these compounds by adding the right amount of this additive to the formulation. This is mainly decided by the loading of silica in the rubber. There is no further advantage gained in mixing any more silane than the amounts discovered in this study. It is worth noting that mixes with 30 p.p.h.r. and 60 p.p.h.r. silica, gain the most benefit from the presence of silane.

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