

# Interface and Mechanical Properties of Peroxide Cured Silicate Nanofiber/Rubber Composites

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**ABSTRACT:** Four representative rubber matrices with varying molecular polarity, such as SBR, NBR, EPDM, and CNBR, were reinforced by mechanical blending with  $\gamma$ -(methacryloxy)propyltrimethoxy silane (KH570) silanized natural fibrillar silicate (FS)/unmodified FS when dicumyl peroxide was used as a curing agent. The effects of the loading amount of silane coupling agent KH570 and the type of rubber matrix on the dispersion of FS as well as the FS-rubber interfacial bonding were investigated. The mechanical properties of various rubber composites filled with unmodified FS and silanized FS individually were compared. As indicated by SEM and TEM, unmodified FS showed better dispersion in polar CNBR than in nonpolar SBR, whereas the improvement of the dispersion is more for SBR after the silanization of FS, constructing a stronger filler network. Compared with unmodified FS/rubber composites, silanized FS/rubber

composites exhibited a higher interphase crosslinking network as well as a lower loss factor, indicating the improvement of the FS-rubber interfacial bonding. The more the loading amount of KH570 was, the more the FS-SBR bonding was improved. Similarly, the improvement of the FS-rubber bonding was more evident in nonpolar SBR and EPDM after FS was silanized, causing the more increase in mechanical properties, relative to polar NBR and CNBR. In strong molecular polar rubber such as CNBR and NBR, the strong interfacial bonding was achieved even if FS was not silanized with KH570, so that the composites filled with unmodified FS also possessed good mechanical properties. © 2008 Wiley Periodicals, Inc. *J Appl Polym Sci* 110: 262–269, 2008

**Key words:** interface; nanofiber; rubber; fibrillar silicate; composites

## INTRODUCTION

With the continual rising of international crude oil price, inorganic fillers become increasingly important for balancing the properties and cost of polymer materials. Especially for rubber, it is necessary to improve the mechanical properties and processing properties by adding large amount of filler, sometimes even up to 70% mass fraction of rubber. Inorganic filler reinforcing polymer faces two key issues: One is the dispersion of filler; the other is the filler-polymer interfacial bonding. Generally, surface energy of inorganic filler is much higher than that of polymer, which causes them to be incompatible with each other and makes it difficult to form a fine dispersion of filler. Especially when the diameters of particles are very small ( $\leq 1 \mu\text{m}$ ), they incline to agglomerate. It is well known that surface modification of filler can improve its dispersion in polymer as well as the filler-polymer bonding.<sup>1</sup>

Fibrillar silicate (as defined FS) microparticles are a class of hydrated magnesium/aluminum silicate, such as palygorskite, composed of a number of needle-like nanosingle crystals (short nanofibers) with a length of 500–2000 nm and a diameter of 10–30 nm.<sup>2,3</sup> The previous researches focused on the anisotropy of FS-reinforced styrene butadiene rubber (SBR) and acrylonitrile butadiene rubber (NBR), surface modification of FS and its reinforcing mechanism on FS/rubber composites, and the structure and properties of SBR/FS with different direct blending methods.<sup>3–5</sup> It was found out that FS/rubber composites showed a typical stress-strain behavior and the anisotropy of short fiber reinforced rubber (SFRC), and the excellent reinforcement of FS resulted from the separation of FS into lots of nanodiameter short fibers, as well as the strong fiber/rubber bonding especially when the silane coupling agent was used to silanize FS.<sup>3,4</sup> It was also concluded that, among silanized FS with various types of silane coupling agents such as bis(3-triethoxysilylpropyl) tetrasulfide (Si69),  $\gamma$ -(methacryloxy)propyltrimethoxy silane (KH570),  $\gamma$ -aminopropyltriethoxy silane (KH550),  $\gamma$ -glycidylpropyltrimethoxy silane (KH560), and vinyltriacetoxysilane (A151), silanized FS with Si69 had the best reinforcement to SBR when sulfur was used as the vulcanizing agent, whereas silanized FS with KH570 was the best when

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dicumyl peroxide (DCP) was used.<sup>4</sup> Surprisingly, KH570 silanized FS-filled SBR composite vulcanized by peroxide exhibited much high tensile strength and stress at the certain strain. However, the dispersion of FS and the FS-rubber interface in peroxide cured FS/rubber composites was discussed less. Generally, the cured rubber with peroxide possesses better heat-resistant and lower compress set, but the lower strength and the lower elongation at break, relative to the cured one with sulfur crosslinking system.<sup>6–9</sup> The C—C crosslinking bonds originated from peroxide vulcanizing system present the lower stress relaxation because of their higher bond energy and shorter bond length. They first endure the stress and fracture when the rubber vulcanizate is drawn, resulting in increase in the asymmetry of crosslinking network, thus the crosslinking network would break at a lower stress.<sup>6</sup> It was hypothesized that more improvement in the interface between FS and SBR might compensate for the asymmetry of C—C crosslinking network, leading to a better reinforcement of KH570-silanized FS in peroxide cured rubber composite. Although peroxide is used less extensively than sulfur in rubber industry, it is preferentially chosen when excellent heat-resistant or very low compress set for rubber materials are required.<sup>8,9</sup>

In present work, four representative commonly used rubber matrices with varying molecular polarity, such as SBR, NBR, ethylene-propylene ethylidene norbornene terpolymer (EPDM), carboxylated acrylonitrile butadiene rubber (CNBR), were reinforced by mechanically blending with KH570-silanized FS/unmodified FS when DCP was used as a curing agent. In two nonpolar rubbers, SBR has more double bond content (C=C) than EPDM; in two polar rubbers, CNBR has the stronger molecular polarity than NBR because of the introduction of a few carboxyl groups. These differences may influence on the dispersion of FS as well as FS-rubber bonding. Therefore, the dispersion of FS in nonpolar SBR and polar CNBR was characterized by scanning/transmission electron microscopy (SEM/TEM) and Payne effect of the FS/rubber compound. Effect of KH570 loading level as well as rubber matrix on the FS-rubber interfacial bonding was further investigated by analyzing the curing properties and dynamic loss factor of FS/rubber systems. The mechanical properties of the various rubber composites filled with unmodified FS and silanized FS individually were compared.

## EXPERIMENTAL

### Materials

Styrene butadiene rubber (SBR1502) and ethylene-propylene ethylidene norbornene terpolymer

(EPDM4045) were produced by Jihua synthesis rubber factory (China); acrylonitrile butadiene rubber (NBRN220S) was brought from JSR company (Japan); carboxylated acrylonitrile butadiene rubber (CNBR506) was manufactured by Nandi company of Taiwan province (China). FS (1250 mesh) was obtained from Dalian Global Mineral Company (China).  $\gamma$ -(Methacryloxy)-propyltrimethoxy silane (KH570,  $\text{CH}_2\text{C}(\text{CH}_3)\text{COOCH}_2\text{CH}_2\text{CH}_2\text{Si}(\text{OCH}_3)_3$ ) was bought from Nanjing Crompton Shuguang Organosilicon Specialties Co. (China). Dicumyl peroxide (DCP) and other chemical agents were purchased from chemical store.

### Samples preparation

#### Silanization of FS

The 20–50 wt % FS aqueous solution was put into a high-speed agitator and agitated at the speed of 800 rpm to make FS-disaggregated/exfoliated into single crystals. The mixture of modifier KH570/alcohol at the ratio about 1 : 5 (mass fraction) was added slowly and uniformly to the aforementioned solution. After the solution was silanized for 1 h at 90°C, the solvent was removed to get the solid particles, and then the solid particles were ground into powder. The mass fraction of KH570 was 1.25%, 2.5%, 6%, 7.5%, 12.5%, and 15% relative to FS.

#### Composites preparation

Two-roll mill was adjusted to the smallest distance when rubber became fluidic, then unmodified FS or silanized FS was added into the rubber step by step to ensure good dispersion by mechanically mixing. Next, the curing agent was added, thus FS/rubber compound (uncured) was obtained (the compositions as shown in Table I). The aforementioned compound was vulcanized to get the composite via the presser at the pressure of 15 MPa.

## CHARACTERIZATION AND TEST

Curing characteristics of the FS/rubber compounds including scorch time ( $T_{10}$ ), curing time ( $T_{90}$ ), minimum torque ( $M_L$ ), and maximum torque ( $M_H$ ) were determined by a disc oscillating rheometer. The vulcanization temperature was set as 150°C for FS/SBR compound, and 160°C for FS/EPDM, FS/NBR, and FS/CNBR compounds.

TABLE I  
Compositions of Materials

|                                       |         |
|---------------------------------------|---------|
| Rubber (phr)                          | 100     |
| DCP (phr)                             | 0.8–3.0 |
| Unmodified FS (or silanized FS) (phr) | 40      |

Payne effect of FS/rubber compound was investigated by RPA 2000 from Monsanto Company (USA). The test frequency was 1 Hz, and the temperature was 60°C. Similarly, the loss factor of FS/rubber composite at different strains was also carried out after the compound was vulcanized in test die at the set temperature and then was cooled down to 60°C.

Fourier transform infrared spectrum Nicolet 210 produced by Nicolet company (USA) was used to study the reactivity of KH570 and peroxide.

A CAMBRIDGE S-250MK3 SEM was used to observe the tensile fracture surface morphology of the composites. A H-800 TEM was used to observe the dispersion of FS in rubber. The ultrafine film for TEM observation was cut by a microtome at -100°C.

Tensile test, tear test, and hardness of composites were carried out by using an Instron tensile machine (Instron1185, USA) according to ASTM D 412, ASTM D 624, and ASTM D 2240, respectively. During tensile test and tear test, five specimens were required to give the average value. During the hardness test, three different spots were measured to give the average hardness value.

## RESULTS AND DISCUSSION

### The dispersion of FS

Figure 1 displays TEM and SEM micrographs of unmodified FS or KH570-silanized FS filled SBR and CNBR composites, which were vulcanized by DCP. From TEM micrographs in Figure 1(a,c), some of FS microparticles, whether being silanized or not, were exfoliated into short nanofibers in SBR and CNBR matrices by mechanically shearing during direct blending with rubber, although some FS agglomerates were observed on the tensile fracture surface of the composites. It is the fact that the interactions between nanosingle crystals (short nanofibers) among FS particles is relatively weaker than that between two layers of clay particles, resulting from a similar line-line contact and the existence of only weak van der Waals force or hydrogen bond.<sup>10</sup> Another important reason for this is that high shear stress on dispersion associated with high viscosity of rubber matrix is achieved.<sup>11</sup> By SEM observation of the tensile fracture surfaces of composites in Figure 1, it was concluded that, as for the same rubber matrix, silanized FS with 6% KH570 presented better exfoliation and dispersion than unmodified FS, and few FS agglomerates were found on fracture surfaces of the composites. Some bigger voids/holes appeared on the fracture surface of unmodified FS/SBR composites because of the pullout of FS from rubber matrix during breaking, indicating poor interfacial bonding, besides some agglomerates with the size ranges of 0.5–4 μm. Also, it seemed that unmodified FS showed a little better exfoliation and dispersion in CNBR than in SBR,

whereas the dispersion of silanized FS was reversely better in SBR. CNBR with strong molecular polarity showed a good compatibility with FS, which helps to improve the dispersion of FS. On the other hand, CNBR might absorb part of KH570 by the interaction of the carboxyls of CNBR and Si—OH of KH570 after hydrolysis, weakening the silanization efficiency of KH570 on FS.

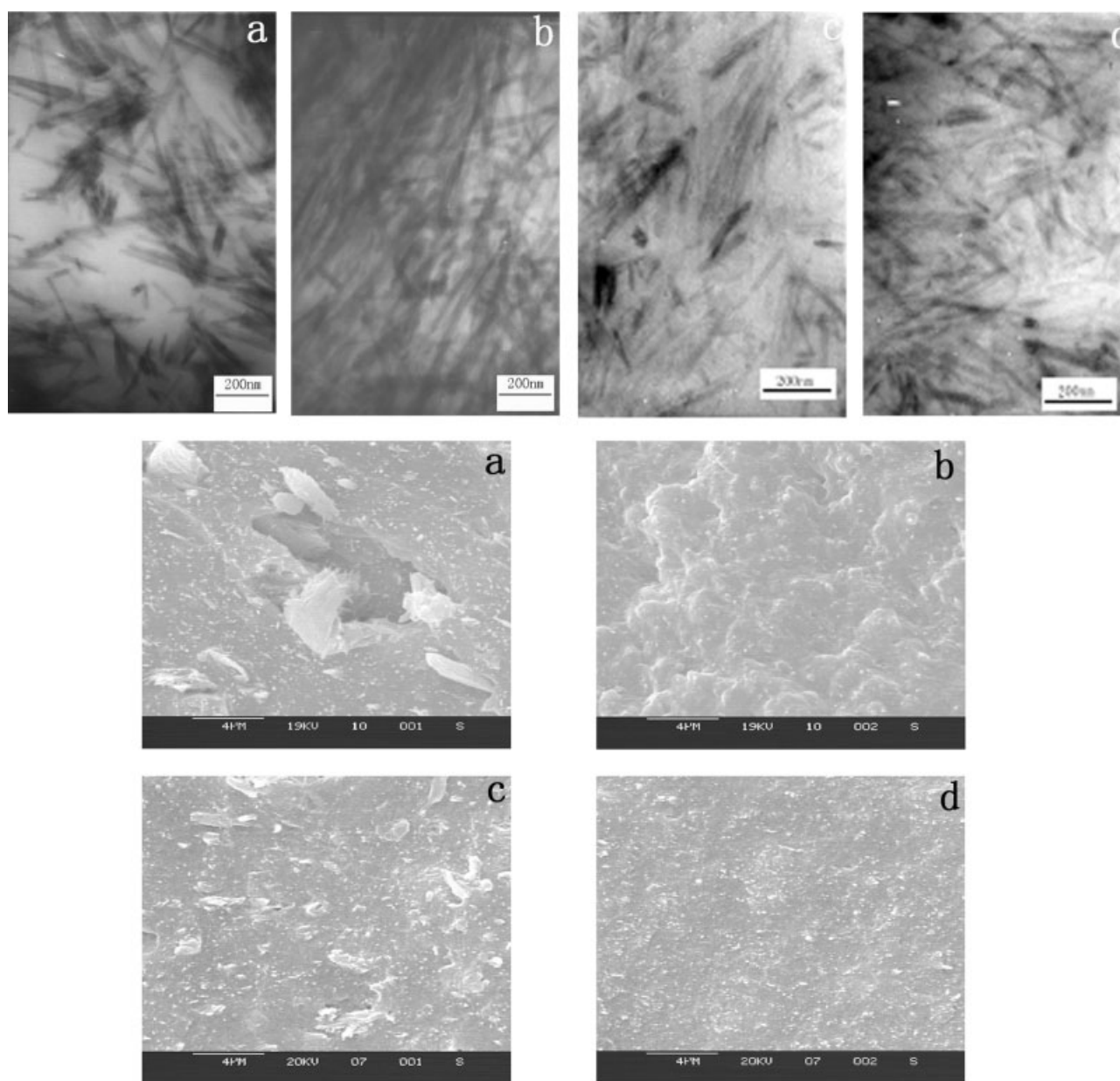
Generally, the network of filler in polymer is characterized by Payne effect,<sup>12–15</sup> which can indicate the dispersion of filler in polymer. For filler-rubber system, the higher initial dynamic storage modulus ( $G'$ ) and the quicker  $G'$  attenuation with the strain reflect the stronger filler network and Payne effect as well. As for FS, FS microagglomerates were first exfoliated into lots of short nanofibers, and then these nanofibers constructed the filler network. At the same FS loading, the better the exfoliation and the dispersion of FS was, the more the number of nanofibers was, resulting in a stronger filler work and Payne effect. To eliminate the contribution of the rubber crosslinking network to dynamic storage modulus, Payne effect of the uncured filler/rubber system (so-called compound) was often used to characterize the filler network. Figure 2 reveals Payne effects for FS/SBR (a) and FS/CNBR (b) compounds filled with unmodified FS and silanized FS, respectively. Whether in SBR or in CNBR, silanized FS/rubber compounds showed stronger Payne effect than unmodified FS/rubber compounding at the same FS loading, implying the better exfoliation and dispersion of silanized FS. However, compared with FS/SBR compounds, the difference in Payne effects between FS/CNBR compounds filled with unmodified FS and silanized FS, respectively, seemed smaller. This was consistent with the earlier TEM and SEM observations.

### Interfacial bonding between FS nanofibers and rubber

The interfacial bonding between FS nanofibers and rubber was indicated by analyzing the curing properties of FS/SBR system with various mass fractions of KH570 and a dynamic loss factor of FS/rubber composites filled with unmodified FS and silanized FS, respectively.

#### Effect of silane coupling agent

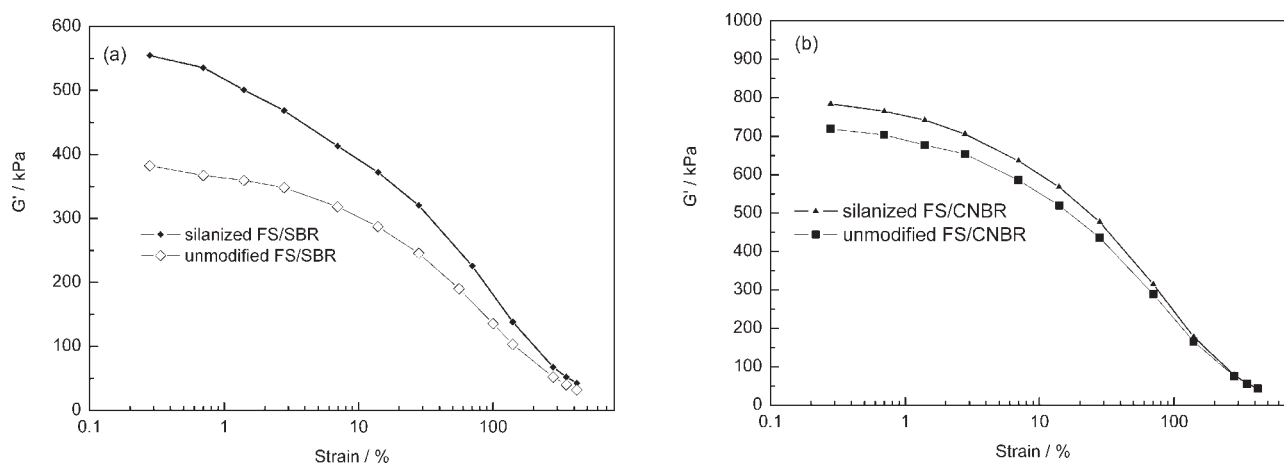
For rubber materials, the curing characteristics at a set temperature were usually measured by using a disc oscillating rheometer, including minimum torque ( $M_L$ ), maximum torque ( $M_H$ ), scorch time ( $T_{10}$ ), and curing time ( $T_{90}$ ). Minimum torque ( $M_L$ ) is related with shear modulus of the compound (before being cured), mainly resulting from the



**Figure 1** TEM and SEM micrographs of FS/rubber composites [(a) unmodified FS/SBR, (b) modified FS/SBR, (c) unmodified FS/CNBR, (d) modified FS/CNBR].

contributions of rubber macromolecular interactions and filler interactions; whereas maximum torque ( $M_H$ ) depends on shear modulus of the composites (after being cured), which is attributed to the effect of the rubber self-crosslinking network and rubber-filler interphase crosslinking network besides the aforementioned two interactions. Therefore, the torque difference ( $\Delta M$ ) between maximum torque ( $M_H$ ) and minimum torque ( $M_L$ ) of the FS/rubber system indicates the difference in shear modulus between the compound and the composite, mainly relying on the contribution of the crosslinking network, including the rubber crosslinking network and rubber-filler interphase crosslinking network as well.<sup>16</sup>

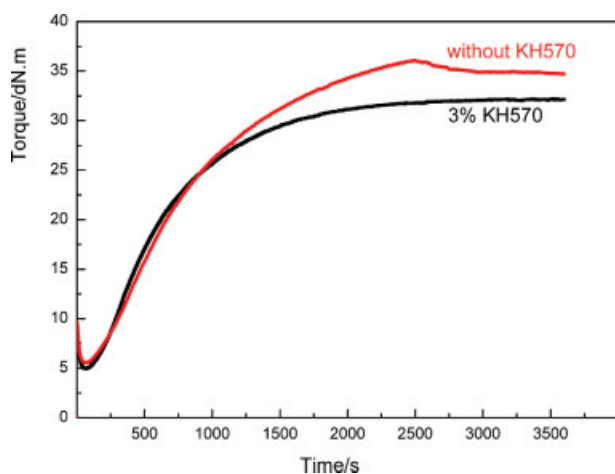
Here the curing properties of neat SBR (unfilled with FS) with and without the introduction of KH570 were first analyzed to reveal the effect of KH570 on the rubber crosslinking network, as shown in Figure 3. Compared with neat SBR, neat SBR with the addition of KH570 showed the lower maximum torque and almost the same minimum torque. This demonstrated that the addition of KH570 decreased the crosslinking network of rubber in that the filler-rubber interphase crosslinking network did not occur due to the absence of FS. It was hypothesized that some DCP was consumed by initiating silane coupling agent KH570. Accordingly, the IR spectra experiment was done to prove the



**Figure 2** Payne effects for FS/SBR (a) and FS/CNBR (b) compounds filled with unmodified FS and silanized FS, respectively.

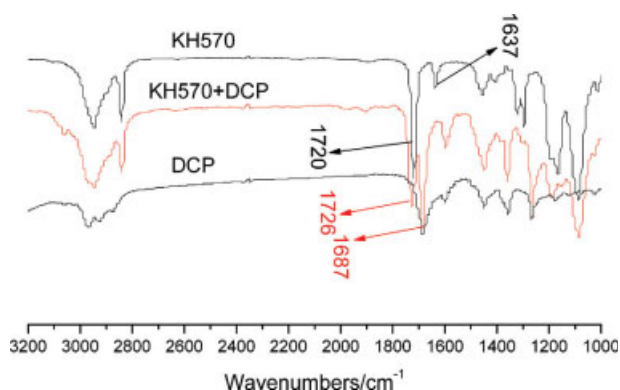
reaction between KH570 and DCP, as shown in Figure 4. For KH570, the absorption peak at  $1720\text{ cm}^{-1}$  was attributed to the stretching vibration of ester  $\text{C}=\text{O}$ , and the absorption peak at  $1637\text{ cm}^{-1}$  was attributed to the stretching vibration of  $\text{C}=\text{C}$ . After the mixture of DCP and KH570 was kept for 15 min at  $160^\circ\text{C}$ , the absorption peak of  $\text{C}=\text{C}$  disappeared; meantime, the stretching vibration of ester  $\text{C}=\text{O}$  at  $1720\text{ cm}^{-1}$  shifted to  $1726\text{ cm}^{-1}$ . DCP was decomposed into methyl free radicals and ketone  $\text{C}=\text{O}$  at  $1687\text{ cm}^{-1}$  while being heated, and these methyl free radicals could initiate the  $\text{C}=\text{C}$  of KH570, causing the disappearance of the  $\text{C}=\text{C}$  absorption peak accompanying with the shift of the stretching vibration of  $\text{C}=\text{O}$ .

Figure 5 presents the torque differences ( $\Delta M$ ) of FS/SBR systems with various amounts of KH570. As seen from Figure 5,  $\Delta M$  of the FS/SBR systems increased with the increasing KH570, indicating the

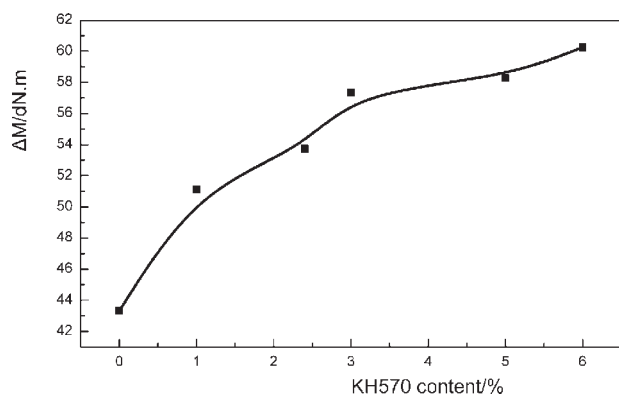


**Figure 3** Curing properties for neat SBR with and without silane coupling agent by peroxide. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

increasing gross crosslinking network. Since the rubber crosslinking network decreased at the moment when KH570 was added into SBR, the increase of gross crosslinking network was attributed to the rubber–filler interphase crosslinking network. Figure 6 shows scorch time ( $T_{10}$ ) and curing time ( $T_{90}$ ) of FS/SBR systems with various amounts of KH570.  $T_{10}$  and  $T_{90}$  for FS/SBR systems shortened with the increasing KH570 loading. There are many of hydroxyls on the surface of FS.<sup>4,17</sup> As analyzed in Ref. 4 the  $\text{Si}-\text{OH}$  of KH570 after hydrolysis may react with the hydroxyls on the surface of FS to form  $\text{Si}-\text{O}-\text{Si}$  bond; on the other hand, DCP produces free radicals by being heated to more than  $130^\circ\text{C}$ , which causes chemical reaction between  $\text{C}=\text{C}$  in KH570 and  $\text{C}=\text{C}$  in SBR. Thus the interphase crosslinking network was acquired. Naturally, the interphase crosslinking network is the main contributor for the modulus of the composites because of the much stronger stiffness of FS relative to rubber macromolecules. As a result, the gross



**Figure 4** IR spectra of KH570, DCP, and DCP/KH570 mixture at  $160^\circ\text{C}$  for 15 min. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

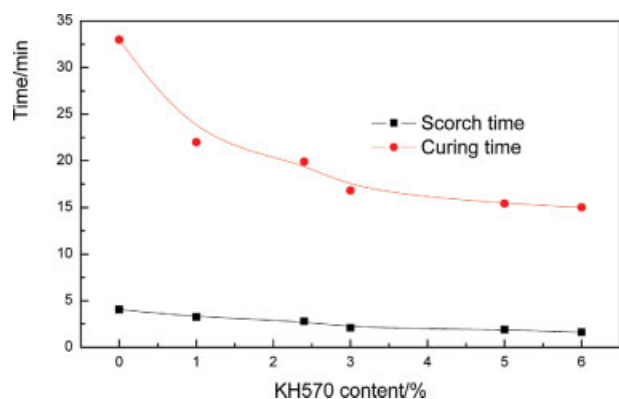


**Figure 5** The torque difference ( $\Delta M$ ) of FS/SBR systems with various amounts of KH570.

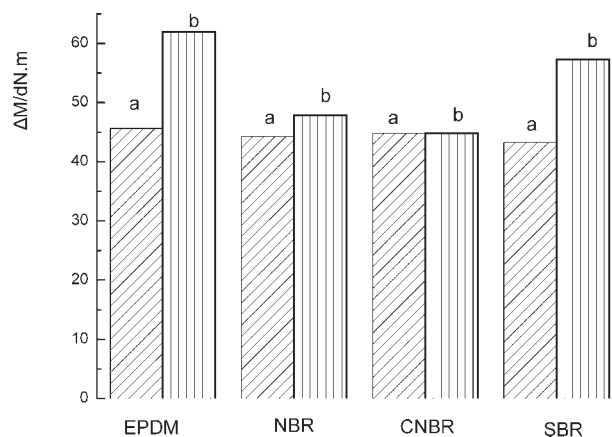
crosslinking degree was increased, and the crosslinking rate was speeded up. It was possible that this increase was slowed down even stopped, if more KH570 was added, because multilayer coating of KH570 might be formed on the surface of FS.

Effect of rubber matrix

Figure 7 depicts the torque difference ( $\Delta M$ ) of various rubber matrices filled with unmodified FS (a) and silanized FS (b), respectively. From Figure 7, compared with the counterpart filled with unmodified FS,  $\Delta M$  for silanized FS-filled SBR and EPDM systems were markedly increased, indicating the increase in the interphase crosslinking network. Whereas, filled NBR and CNBR systems changed little. As shown in Table II,<sup>18</sup> nonpolar EPDM and SBR have lower surface energy, and so the affinity between FS and them is worse in case of free KH570. The interphase crosslinking network for these two nonpolar rubbers was drastically improved by the silanization of FS with silane coupling agent KH570. As for FS/NBR and FS/CNBR systems, this silanization effect was markedly



**Figure 6** Scorch time and curing time of FS/SBR systems with various amounts of KH570. [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]



**Figure 7** The torque difference ( $\Delta M$ ) of various rubber matrices filled with unmodified FS (a) and silanized FS (b), respectively.

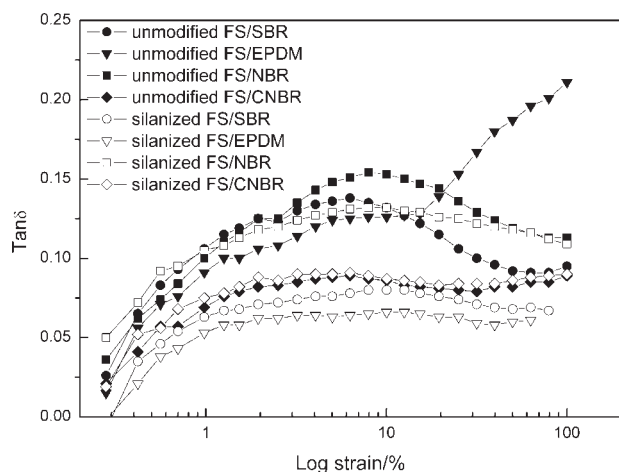
weakened. Both NBR and CNBR with strong polar group have higher surface energy, producing the dipolar effect between FS and them, and at the meantime the hydrogen bond could be formed between the cyanogen group in NBR/carboxyl in CNBR and the hydroxyls on the surface of FS. These two effects remarkably improved the affinity or even the rubber-filler interfacial bonding, despite FS was not silanized with KH570. Even if the chemical interfacial bonding could be formed instead of the earlier physical effects in the aforementioned two polar rubber matrices when FS was silanized with KH570, the number of interphase crosslinking network increased little.

Loss factor ( $\tan \delta$ ) is another indicative of interfacial bonding between filler and polymer.<sup>19</sup> Figure 8 discloses the dependence of loss factor of various rubber composites on shear strain filled with silanized FS and unmodified FS individually. As seen from Figure 8, loss factor ( $\tan \delta$ ) for all the silanized FS/rubber composites presented lower  $\tan \delta$  compared with the corresponding unmodified FS/rubber composites at the tested shear strain. It was also concluded that the loss factor difference for SBR or EPDM filled with silanized FS and unmodified FS, respectively, was bigger, whereas it was the smaller for CNBR or NBR systems. This implied that as for nonpolar rubber such as SBR and EPDM, the interfacial bonding was more remarkably improved after FS was silanized, compared with polar rubber such as NBR and CNBR. This agreed with the discussions mentioned earlier. Furthermore,

**TABLE II**  
Surface Energy of Various Rubbers

| Rubber | Surface energy at 20°C (J/m <sup>2</sup> ) |
|--------|--|
| EPDM   | 34.5                                       |
| SBR    | 40   |
| NBR    | 52.6 <sup>a</sup>                          |

<sup>a</sup> With 27 wt % cyanogens group.

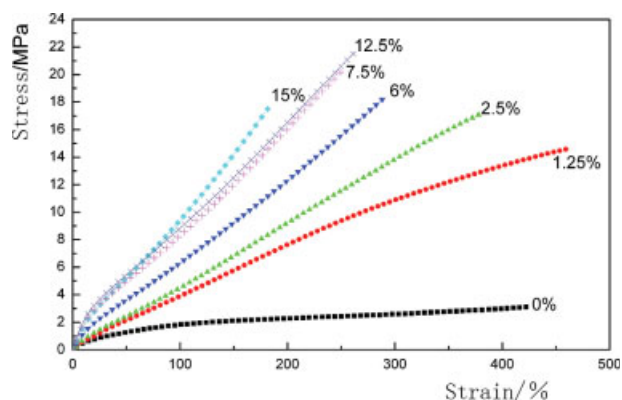


**Figure 8** The dependence of dynamic loss factor ( $\tan \delta$ ) of various rubber composites on shear strain filled with silanized FS and unmodified FS individually.

$\tan \delta$  continued rising with the increasing strain after the maximum value, except that the unmodified FS/EPDM composite always kept increasing because of poor interfacial bonding.  $\tan \delta$  of the composite includes the contributions of rubber macromolecules, rubber–filler interactions, and filler–filler interactions, so it is not comparable among  $\tan \delta$  of the various rubber-based composites. At 60°C, much higher than the glass transition temperature of rubber, rubber exhibited excellent entropy elasticity and very low loss factor. At the same rubber content, the loss factor of the composites was mainly attributed to filler–filler interactions at the lower strain.<sup>20,21</sup> After the appearance of the maximum  $\tan \delta$ , the loss factor continued rising mainly resulted from the relative motions of filler–rubber macromolecules at the higher strain (the effect of interfacial bonding). Strong interfacial bonding could effectively obstruct the change of filler network as well as the relative motions of filler–rubber macromolecules, lowering the loss factor of the composites.

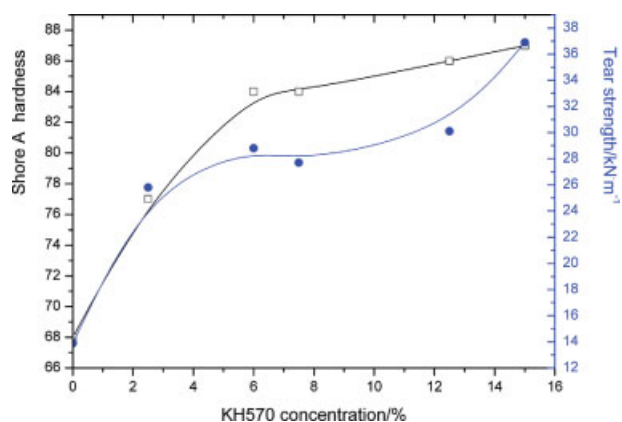
### Mechanical properties

From Figures 9 and 10, compared with unmodified FS/SBR composites, silanized FS/SBR composites exhibited a considerably sharper stress-up with the strain and higher stress, especially at the very low strain, hardness, and tear strength, similar to that of microshort fiber-reinforced rubber composites (SRFC) filled with short nylon fiber, short polyester fiber, short cellulose fiber, and so on.<sup>22–24</sup> As discussed afore, silanized FS had a better dispersion and much improved the interfacial bonding, resulting in the better reinforcement. As the amount of silane coupling agent KH570 increased, this reinforcement was improved. With the further increasing of KH570, the improvement in mechanical properties of silanized FS/SBR composites was rather little.

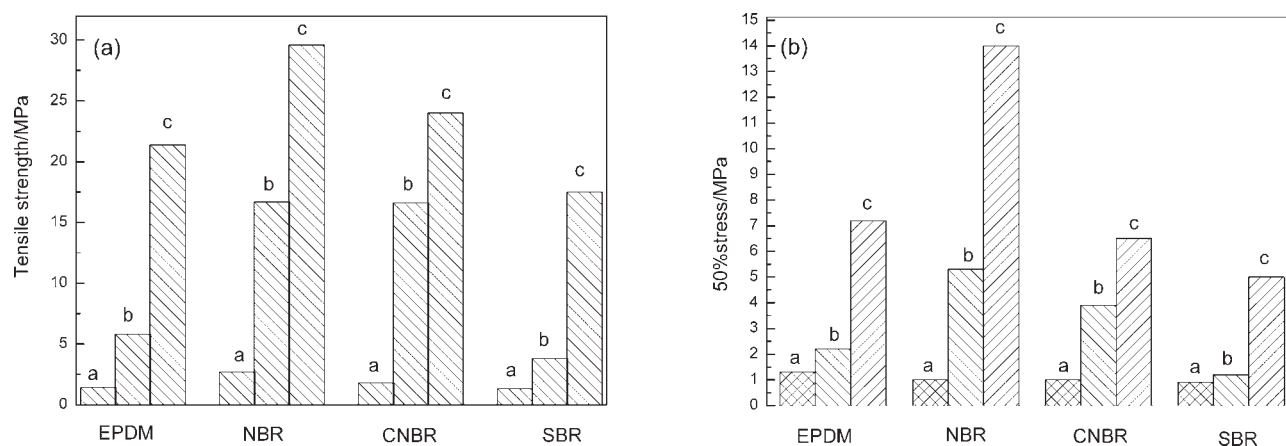


**Figure 9** Stress strain behavior of silanized FS/SBR composites with various KH570 amounts (mass fraction). [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]

Figure 11 displays the comparison of tensile strength and stress at 50% strain of neat rubber (a) and the corresponding composites filled with unmodified FS (b) and silanized FS (c) individually. From Figure 11, stress at 50% strain and tensile strength of pristine rubber vulcanizates, such as SBR, EPDM, NBR, and CNBR, were pretty low. Compared with pristine rubber vulcanizates, these composites, which were prepared by incorporating FS into the corresponding rubber, exhibited higher stress at 50% strain and tensile strength. Evidently, these composites filled with silanized FS got the most distinct improvement. To be noted, relative to unmodified FS/rubber composites, the stress at 50% strain for silanized FS/SBR and silanized FS/EPDM composites increased by 420 and 330%, and tensile strength increased by 460 and 369% individually; whereas the stress at 50% strain for silanized FS/NBR and silanized FS/CNBR composites rose by only 270 and 170%, and the tensile strength rose by 177 and 145%, respectively. In strong molecular



**Figure 10** The hardness and tear strength of silanized FS/SBR composites with various KH570 amounts (mass fraction). [Color figure can be viewed in the online issue, which is available at [www.interscience.wiley.com](http://www.interscience.wiley.com).]



**Figure 11** The comparison of tensile strength and stress at 50% strain of neat rubber (a) and the corresponding composites filled with unmodified FS (b) and silanized FS (c) individually (6% KH570).

polar rubber such as CNBR and NBR, even if FS was not silanized with KH570, the composites filled with unmodified FS still possessed good mechanical properties. For example, tensile strength and the stress at 50% strain for unmodified FS-filled NBR and CNBR composites still reached 16.7 MPa and 5.3 MPa, 16.3 MPa and 3.9 MPa, individually. These differences in reinforcement were due to the difference in both the dispersion of FS and the FS-rubber interfacial bonding in various rubber matrices. As aforesaid, the silanization of FS could more remarkably improve the dispersion of FS as well as the FS-rubber interfacial bonding in nonpolar SBR and EPDM than that in polar NBR and CNBR, causing the more visible increase in mechanical properties for nonpolar SBR and EPDM. In strong molecular polar rubber such as CNBR and NBR, the strong interfacial bonding was achieved by the good compatibility of FS and polar rubber in case of free KH570.

## CONCLUSIONS

When peroxide was used as the curing agent, the silane coupling agent KH570 and the type of rubber matrix both influenced the dispersion of FS as well as the FS-rubber interfacial bonding. Unmodified FS showed better dispersion in polar CNBR than in nonpolar SBR. The improvement of the dispersion is more for SBR after the silanization of FS, leading to a stronger filler network. Compared with unmodified FS/rubber composites, silanized FS/rubber composites exhibited the higher interphase crosslinking network as well as the lower loss factor, indicating the improvement of the FS-rubber interfacial bonding. The more the loading amount of KH570 was, the more the FS-SBR bonding improved. Similarly, the improvement of the FS-rubber bonding was more evident in nonpolar SBR and EPDM after FS was silanized, causing the increase in mechanical properties, relative to polar NBR and CNBR. In strong molecular polar rubber such as

CNBR and NBR, the strong interfacial bonding was achieved even if FS was not silanized with KH570, so that the composites filled with unmodified FS also possessed good mechanical properties.

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