

Thermal and Mechanical Behavior of Natural Rubber Latex-Silica Aerogel Film

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ABSTRACT: A major problem in most natural rubber latex (NRL) commonly encountered like other polymer is susceptibility to mechanical properties and thermal degradation; particularly in thin film due to the presence of double bonds in the main chain. Therefore, it is desirable to seek for ways of improving these properties. Silica aerogel is a material with extraordinary properties was believed to have potential enhance properties in NRL films because of its high specific surface area. Therefore, based on the unique character of silica aerogel, NRL-silica aerogel film was developed by latex compounding and dry coagulant dipping to form thin film where silica aerogel acts as filler. Silica aerogel, synthesized from rice husk

was dispersed in a ball-mill using distilled water for NRL compounding. Results indicate that increasing silica aerogel loading enhances the mechanical properties of the NRL-silica aerogel film. Effects of postvulcanization processes were also investigated, whereby the best reinforcing effect was obtained at 4 phr silica aerogel loading with leaching postvulcanization condition. © 2011 Wiley Periodicals, Inc. *J Appl Polym Sci* 124: 3108–3116, 2012

Key words: natural rubber latex-silica aerogel film; high temperature rubber composite; mechanical susceptibility; thermal susceptibility

INTRODUCTION

Natural rubber latex (NRL) gives excellent chemical and physical properties as natural elastomer. It contains mainly unsaturated *cis*-1,4-polyisoprenes which can be manufactured via a relatively safe process. Commonly, NRL encounters a major problem like any other polymer, which is susceptible to oxidation or thermal degradation and low mechanical strength, particularly when formed into thin films.¹ In addition, natural rubber products can biodegrade by a combination of chemical and biological reactions. In recent years, synthetic rubbers are largely manufactured because it possesses better chemical and physical properties compared to natural rubber.² However, synthetic rubbers are not environmental friendly and the production of which often involves use of hazardous chemicals.³

A huge majority of latex gloves used by healthcare professionals, emergency medical technicians, police officers, fire personnel and chemists are made of NRL. Therefore, natural rubber needs to be rein-

forced and cured by fillers such as carbon black, ultra-fine calcium carbonate, modified montmorillonite, starch, and silica.^{1,4–6} Reinforcement in NRL is primarily the enhancement of strength and strength-related properties, abrasion resistance, hardness, tensile strength, thermal stability, and modulus.^{1,5,7,8} Good reinforcing filler should have chemically active surface which is both porous and very irregular in shape, in order to maximize surface contact between rubber and filler.⁷ Properties of the filled rubber depend mainly on the physical and chemical interactions at the interface of rubber-filler surface. Mechanical properties like ageing resistance, tear resistance, tensile strength, elongation at break, and thermal properties of the NR composite is moderate because of the self-aggregation of the fillers due to the supersaturation and interaction density.^{1,7,9} The ability to tailor the mechanical and thermal properties of latex articles is of great interest and has many potential applications.

Silica has been widely used as the main reinforcing filler that increases the usefulness of rubber by increasing the tensile strength, tensile modulus as well as tear strength of the resulting rubber products.⁴ Overloading of silica tends to reduce the strength of rubber due to aggregation issue. It was observed that good dispersion of silica improved the thermal and mechanical properties of rubber composite. However, the loading must be controlled because as more silica is loaded, heavy secondary

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aggregation of silica is gradually generated resulting in loss in mechanical advantage.⁷ It has already been underlined in many references that the particle size, specific surface area, and surface activity are crucial parameters in deciding the interaction between rubber matrix and filler.¹⁰ A study showed that, low surface area filler gives low chemisorbed bound of rubber.¹¹ The high reinforcing effectiveness of composites at low loading of layered silicates is due to the nanoscale dispersion and very high aspect ratio of the silicate platelets. Therefore, using fillers with high specific surface area and small size such as spherical inorganic nanoparticles to directly reinforce NR has the potential to improve the material properties. An example of such material is silica aerogel.

Silica aerogel is a nanostructured material with high specific surface area, high porosity, low density, and excellent heat insulation properties.⁹ Silica aerogel was initially made from tetramethylorthosilicate (TMOS) or tetraethylorthosilicate (TEOS) as the precursor.^{1,6} It was then replaced by less corrosive and expensive precursor such as water glass or sodium silicate.⁴ Recently, silica aerogel was synthesized from rice husk ash (RHA) as an efficient source of silica via a green process.⁶

Current environmental issues which are common worldwide are global warming, green house effect, and climate change. Awareness of this environmental concern has led to the rise in effort to renew agricultural waste. In the case of rice husk, disposal of the waste has become a problem because open burning is normally practiced. As a consequent of the burning process, about 20 wt % of the RHA residues is produced; causing environmental problems such as severe air contamination and water pollution. On the other hand, controlled combustion of rice husk can provide a pure source of silica which is a highly useful raw material for a variety of applications, including the synthesis of silica aerogel.^{4,6}

In this article, we report on the study on thermal and mechanical properties of NRL reinforced with silica aerogel. The objectives of the study are to compound silica aerogel in NRL films, study the effect of post vulcanization processes on NRL-silica aerogel films and determine the thermal and mechanical properties of these films compared with other different types of fillers.

EXPERIMENTAL

Materials

NRL was purchased from Zarm Scientific & Supplies (Malaysia) Pte. Dry rubber content and total solid content of the NRL were 60.07% and 61.5%,

respectively. Mechanical stability time of NRL was 1560s and volatile fatty acid number was 0.026. Silica aerogel was prepared in the laboratory while rice husk was obtained from a local paddy field. Silica aerogel filler dispersion was prepared by grinding the silica aerogel powder and dispersing ingredients in a ball mill for 24 h. The dispersing ingredient was distilled water (90% wt/wt). The compounding material, zinc oxide, zinc diethyldithiocarbamate (ZDEC), sulphur, antioxidant, and Vulkasil C were obtained from Farben Technique, Malaysia. The compounding ingredients in dispersion state being premixed in separated beaker to ensure homogenization of the compounding ingredients. All compounding materials were used as received.

Synthesis of silica aerogel from rice husk

Silica aerogel was synthesized according to established procedure by Hamdan.⁶ Rice husk was calcined at 700°C to obtain RHA. Subsequently a mixture of RHA (39.13 g), NaOH (14.55 g), and H₂O (450 g) was stirred in a Teflon bottle at 90°C for 2 days and the mixture was then filtered. The obtained filtrate is known as sodium silicate. Gelation took place in a few minutes time after 13 mL of catalyst, sulphuric acid was added into the sodium silicate solution, followed by aging for 48 h at room temperature. The aquagel was washed in distilled water until the pH of the aquagel was adjusted to about 6 and filtered to remove soluble salts. The aquagel was then converted into alcogel by solvent exchange process using soxhlet extraction. Finally, silica aerogel was prepared by supercritical drying of alcogel in ethanol at 239°C and 1200 psi, respectively, using Parr instrument autoclave fitted with thermocouple, a pressure gauge and a temperature controller. Finally, the silica aerogel was collected and kept in a Teflon bottle.

Preparation of pre vulcanized NRL films

Silica aerogel synthesized from rice husk was dispersed in ball-mill for 24 h using distilled water in order to render it compatible for latex compounding. Table I shows the TSC and compounding formulation. NRL-silica aerogel films were developed through latex-compounding techniques. Typically, all ingredients including the silica aerogel filler were mixed simultaneously and stir for 1 h at 270 rpm with mechanical stirrer. The mixture was then pre vulcanized at 80°C until chloroform number 3 is achieved and leave for maturation for 24 h at room temperature.⁹ Upon compounding, the NRL-silica aerogel films were prepared through the dry coagulant dipping. Dipping plates were cleaned and dipping procedures were done by dipping the plates in

TABLE I
Compounding Formulation of NRL-Silica Aerogel Film

Ingredients (%)	Dry weight (phr)
61.1 NRL	100
10.0 Potassium hydroxide	0.3
51.3 Sulfur	0.5
53.4 ZDEC	0.75
33.1 Zinc Oxide	0.25
52.2 Antioxidant 2246	0.5
10.0 Silica aerogel	0, 1, 2, 3, 4, 5, and 6

coagulant tank for 13 s, dried in oven for 5 min at 80°C before dried at room temperature for 2 min. The plates were then dipped into the prevulcanized NRL compound for 17 s and curing in the oven for 20 min at 100°C. The formed films were stripped off from the plates by the help of Calcium Carbonate and stored in a refrigerator before further testing. Leaching was done with soaking the specimens into a beaker with distilled water for 24 h, while aging was done by heating the specimens in oven at 100°C for 3 days.

Characterization and testing

The surface area of fillers was determined by means of BET N₂ adsorption using Qsurf surface area analyzer (Thermo Finnigan, Italy). The analyzer was calibrated using Al₂O₃ as the standard material prior to analysis.

Fourier Transformed Infrared Spectroscopy (FTIR) spectra of the samples were recorded by Alpha FTIR Spectroscopy (Bruker Optics, MA, USA) for samples functional groups.

The mechanical behavior of the rubber materials were conducted with Instron Universal IX Automated Materials Testing System (Instron, Acton, MA). For this purpose the specimens were cut from molded sheets in the dumb bell shape according to the ASTM D-412 for tensile strength and, ASTM D-624 for type C tear strength both with cross head speed of 500 mm/min at room temperature. All of the mechanical property values were obtained by averaging the five experimental values.

Morphology of silica aerogel was taken with Field emission scanning electron micrograph (FESEM) using JOEL, JSM-6701F. A sputter coater was used to coat sample with platinum to avoid charging under the electron beam.

NRL film was observed with scanning electron micrographs (SEM) using (JOEL, JSM-6390LV) and transmission electron micrographs (TEM) using (FEL, Tecnai G2 Series). The samples were quenched in liquid nitrogen bath and split under freezing condition. The samples were also coated with platinum to avoid charging.

The thermal stability of the samples from 30 to 600°C was investigated with a Perkin Elmer Pyris 6 thermogravimetric analyzer, TGA (Perkin Elmer, MA, USA) instrument at a heating rate of 10°C/min in nitrogen atmosphere.

The glass transition temperature was measured by Mettler Toledo DSC 1 calorimeter over the temperature range of -100 to 70°C at a heating rate of 10°C/min under nitrogen atmosphere. The NRL thin films samples (c.a. 10 mg) were encapsulated in standard, aluminum pans. The samples were pre-treated at 200°C at a rate of 20°C/min for 1 min at this temperature in order to remove the volatile impurities. The samples were quenched to -80°C at a rate of 10°C/min. The DSC scan was made from -80 to 70°C at a heating rate of 10.00°C/min in nitrogen atmosphere. The glass transition temperature, T_g of each sample was taken as the midpoint of the transition.

RESULTS AND DISCUSSION

Specific surface area of silica aerogel

Brunauer-Emmett-Teller (BET) specific surface area of the different filler type is shown in Table II with silica aerogel having a surface area of 402 m²/g. The increase of modulus and tensile strength is very much dependent on the particle size of the filler; smaller particulate fillers imparting greater reinforcement to the rubber compound than the coarse ones.^{12,13} Since, particle size is directly related to the reciprocal of surface area per gram of filler, it is the increase in surface area that is in contact with the rubber phase which probably leads to the increase in reinforcement.^{10,11,14,15} As the specific surface area of silica aerogel is high, it enhances the properties of NRL thin films.

Morphology of silica aerogel

It is observed that silica aerogel in general has a uniform and spherical structure of less than 50 nm in diameter (Fig. 1). The mean particle size distribution of silica aerogel measured and calculated by SEM shown in Figure 2 is broad, with a major distribution at a low particle size range of 10–30 nm. A smaller distribution of larger particle size range of 40–80 nm indicates agglomeration of nanosilica

TABLE II
BET Surface Area of Different Filler Types

Sample	Surface area (m ² /g) ±5%
Silica aerogel	402.0
Rice husk ash (RHA)	51.4
Vulkasil C	55.0

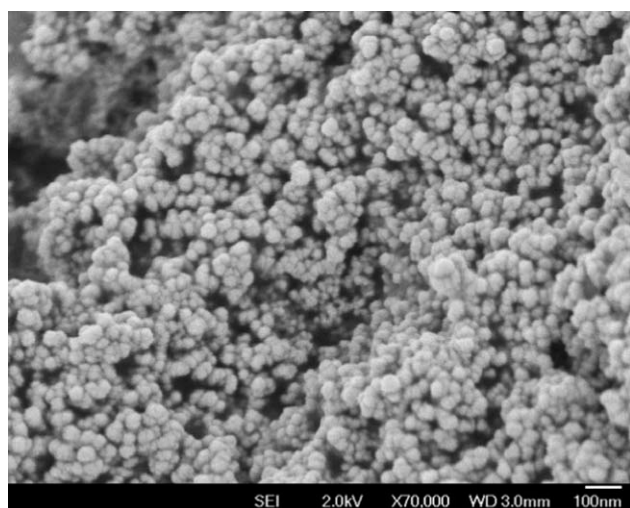


Figure 1 FESEM image of silica aerogel made from rice husk ash.

particles. Low particle size simply results in a greater influence of polymer-filler interaction. In addition to average particle size, the particle-size distribution also has a significant effect on reinforcement.¹⁵ Particulate fillers with a broad particle-size distribution have better packing in the rubber matrix, which results in a lower viscosity than that provided by an equal volume of filler with a narrow particle-size distribution. Another important concern in reinforcement is the presence of large particles or agglomerates in the rubber. These agglomerates not only reduce the contact between filler and matrix but function as failure initiation sites which would lead to premature failure of materials.^{11,16} Evidently, it is not a problem for silica aerogel as its surface area and particle size is satisfied.

Mechanism of the NRL filler mixing

When filler particles are dispersed with NRL, the predicted coreshell structure is formed as depicted by the TEM micrograph of NRL/silica aerogel thin

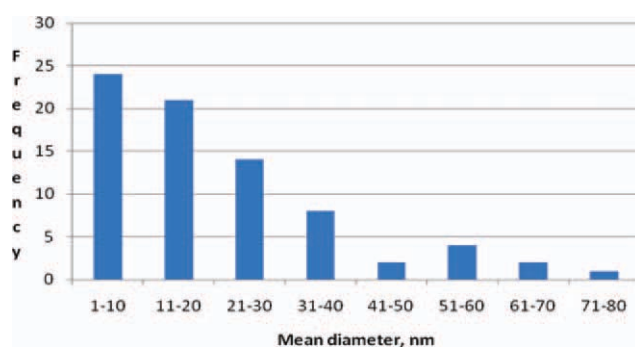


Figure 2 Particles distribution. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

film in Figure 3. The silica-aerogel; appears as the dark image exists as the core or filler, surrounded by the lighter shell structure of the NRL matrix. In addition, for polymer-filler interactions, physical adsorption of rubber molecules may take place on the filler surface.

It is observed that the greater is the specific surface area, the smaller is the particle size. The surface effect seemed stronger, which increased the contact area between the filler and rubber particles and ensured the formation of physical intertwines. Hence, the particle diameter of the reinforcing agent was the main factor for its reinforcing ability. Better diffusion efficacy is observed as the particle diameter gets smaller. In addition, its matching effect with the free volume of rubber tends to be stronger, role as an impurity is weaker and ability to inhibit the micro-crack expansion is higher.²

At the same time, the ability to inhibit the movement of macromolecules and the carrying efficiency increased, resulting in a good reinforcing effect. However, when the amount of reinforcing agent exceeded a critical value, the contact of rubber particles with reinforcing-agent particles became saturated. When the amount of reinforcing agent continued to increase, the aggregates became larger and the distance between the rubber particles increased, which broke down the monolithic construction of the material; reducing the reinforcing effect.

Mechanical properties

Effect of silica aerogel loading

It can be seen that with increasing amount of silica aerogel loading in compounding, tensile strength, modulus and tear strength of the thin films were increased and reached an optimum value when the silica aerogel amount was 4 phr (Figs. 4–6). The

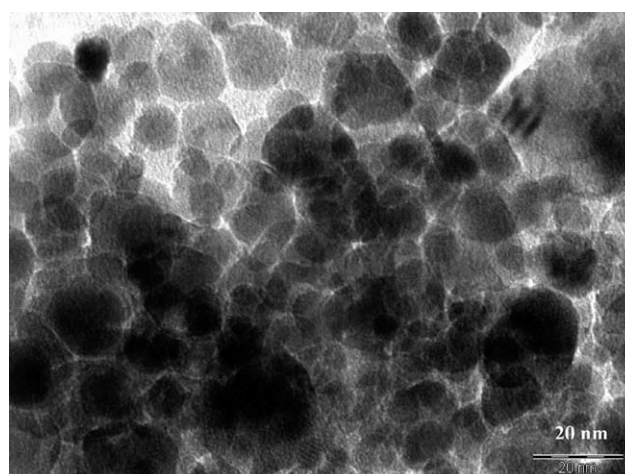


Figure 3 TEM micrograph showing the core-shell structure of NRL/silica aerogel thin film.

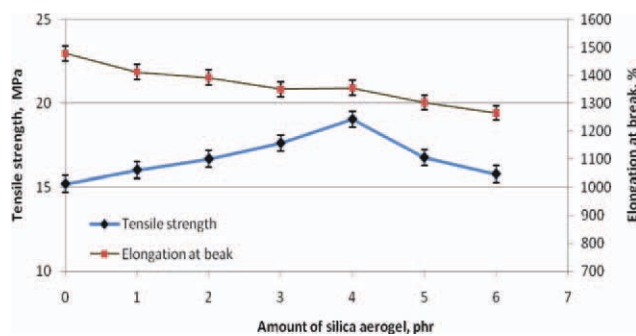


Figure 4 Effect of filler loading on tensile strength and elongation at break of NRL-silica aerogel thin films. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

elongation at break gradually decreased, whereas the modulus of latex film increased (Fig. 4). It is apparent that silica aerogel has a good reinforcing effect on latex film with an optimum comprehensive efficacy when the amount of silica aerogel was 4 phr. However, when it reached 6 phr, reinforcing effect is low.

The greater specific surface area and smaller particle size of silica aerogel tends to strengthen the surface, increase the contact area between the filler and rubber particles and ensure the formation of physical tangles. At the same time, the ability to inhibit the movement of macromolecules and the carrying efficiency increased, resulting in a good reinforcing effect. But when the amount of reinforcing agent exceeded an optimum value, the contact of rubber particles with reinforcing-agent particles becomes saturated. Further increase in the amount of reinforcing agent increased the size of aggregates and distance between the rubber particles, which broke down the monolithic construction of the material, reducing the reinforcing effect. Addition of silica aerogel resulted in an increase in modulus because of filler–filler interaction, where rubber polymer is partially trapped in secondary silica network.¹⁷ There has been considerable evidence supporting

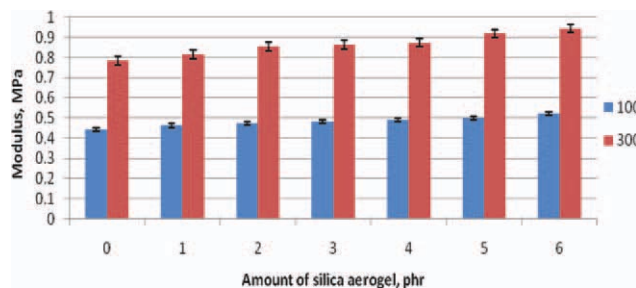


Figure 5 Effect of filler loading amount on modulus at 100% and modulus at 300% of NRL/silica aerogel thin films. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

the tendency of filler aggregates to associate to form agglomerates, especially at high filler loading.^{18,19}

This phenomenon is due to the interaction between filler and rubber particles. Before the addition of filler inside the films, the physical properties of latex are dominantly contributed by formation of sulfur crosslinking which bonds the rubber particles. As silica aerogel filler is added to the system, the film structural integrity became largely dependent on the silica aerogel content.

At low filler content, film integrity is dominated by rubber–rubber interaction. But further addition of filler may also contribute towards the films physical properties. This is seen in Figure 4 which shows that, at 1 phr, the tensile strength of film showed an increased trend. As the filler loading increased, filler–rubber interaction increased until it reached an optimum level. During this transition, the film properties increased gradually with predominant factor shift from rubber–rubber to rubber–filler interactions.

Rubber–rubber interaction is usually dominated by crosslinking formation between the particles, the interactions of the main chain and the Van der Waals forces between the rubber molecules. When the filler is impregnated inside the system, Van der Waals forces are formed between the rubber particles and the filler molecules. Silica aerogel fillers may act as a strengthening medium which supports the rubber particles and reinforced material inside the film. This formation of more interaction may continue until all the rubber and filler particles are fully interconnected.

Further addition of filler beyond the optimum value tends to increase the possibility of filler agglomeration and renders the film become non-aligned during applied stress. In order for the latex film to resist deformation during applied stress, the backbone chain of the macromolecules should be aligned, forming a crystal region that enhanced the films tensile properties. As the filler agglomeration increased, alignment could not be achieved, and

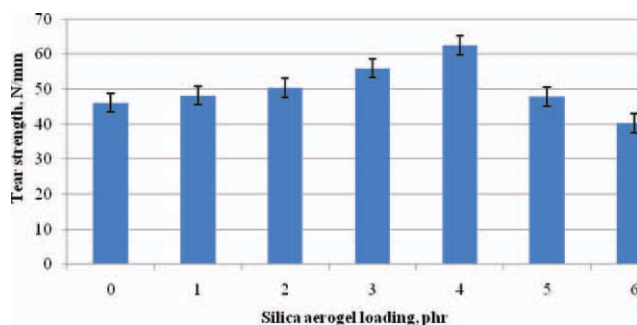


Figure 6 Effect of filler loading amount on tear strength of NRL-silica aerogel thin films. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

thus, the film tends to break easily. Agglomeration sometimes tends to reside between the polymeric backbones chain and the crosslinking formation. Such interstitial agglomeration causes the rubber–rubber and filler–rubber crosslink to expand and stretch until it breaks. This phenomenon reduces the physical properties of the films.

Figure 4 shows that as the filler loading increased the elongation at break for all the films were decreased, due to the fact that addition of filler tends to make the film stiffer. The increasing interaction between the filler and rubber particles tends to increase the crosslink density of the film. Cross-linking tends to increase the entanglements between the backbone chains. Saturation of entanglement limits the mobility of the backbone chain and its ability to align during stretching, instead of providing hindrance towards the film deformation. Consequently, as a stretching force is applied, the film may easily break at lower percentage of elongation. Evidently, filler-polymer interaction play a critical role at a low elongation (100%), while at a high elongation (300%), the strain-reduced crystallization of NR dominates.³ Filler loading has similar effect on both M100 and M300 (Fig. 5), whereby both parameters increase with increasing filler content.^{14,15}

Another significant improvement is observed in tear resistance. Poor tear resistance is one of the major problems for NRL product. Introducing silica into NR can significantly improve its tear resistance. The study indicates that tear strength increases with an increasing silica aerogel loading (Fig. 6). However, there is a significant decrease in tear strength at silica aerogel content of 5 phr, presumably due to severe self-aggregation of silica aerogel.

Study of postvulcanization processes

There are two types of postvulcanization processes tested for its mechanical properties [Fig. 7(a–c)] which are leaching and aging. Optimum condition was observed for film prepared by leaching procedure. Leaching removes extractable protein and impurities on the surface of the thin film; enabled the rubber surface to perform better mechanical strength.

However, for film that was already aged, the mechanical strength decreased because of thermal degradation (breaking of monosulphidic bonds) of rubber chain or backbone chain of the macromolecules (mainly polyisoprene) by oxidation process. During the process, double bonded carbon reacted with oxygen to create epoxide or open structured group, acted as transition group and ended with chain breakage forming two distinct groups.¹ Degeneration of macromolecules chain consequently lowers the strength of the film. The data indicate that quantity

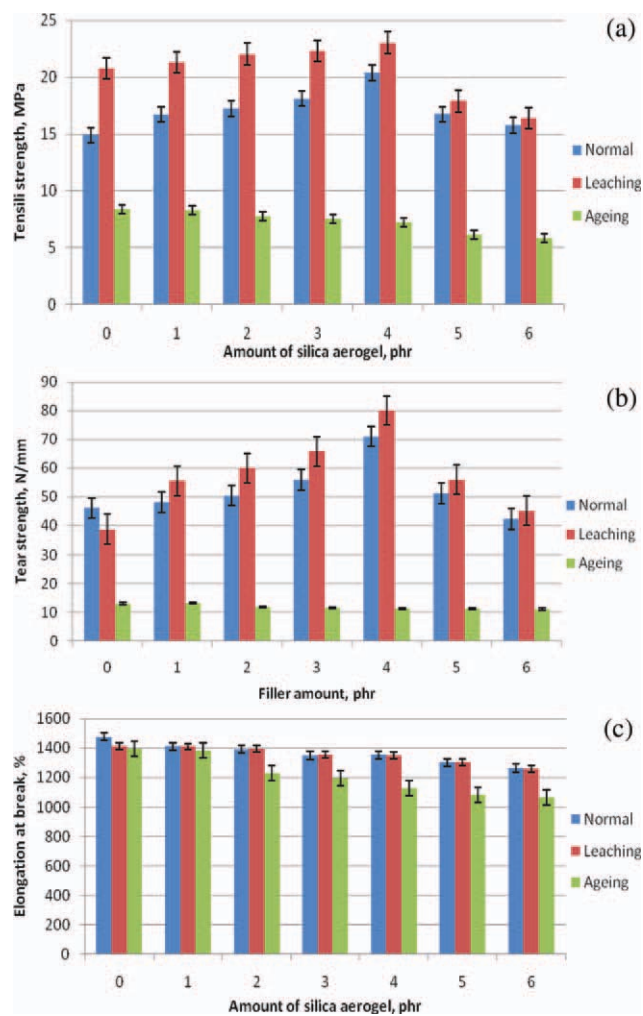


Figure 7 Effect of postvulcanized conditions on (a) tensile strength, (b) tear strength, and (c) elongation at break of NRL-silica aerogel thin films. [Color figure can be viewed in the online issue, which is available at www.interscience.wiley.com.]

of silica aerogel did not have a great effect on mechanical properties of film prepared by aging process.

FTIR study

Figure 8 shows the FTIR spectrum of silica aerogel and NRL films. For silica aerogel, a broad peak appears at 3441 cm^{-1} assigned to O–H stretching due to adsorbed water, H-bridging hydroxyl ($-\text{Si}-\text{OH} \dots \text{O}-\text{Si}-$) group and isolated ($-\text{Si}-\text{OH}$). The peak at 1636 cm^{-1} corresponds to O–H of water. The spectrum also shows strong peaks at around 1107 , 801 , and 490 cm^{-1} due to asymmetric, symmetric, and bending modes of SiO_2 respectively; typical of silica material. The peak at around 1165 and 990 cm^{-1} are assigned to $\text{Si}-\text{OCH}_2\text{CH}_3$ bonds.

Hydrogen bonding between the $-\text{OH}$ groups in the SiO_2 network was studied by observing the shift

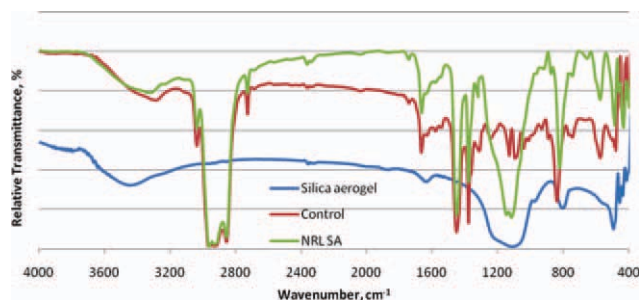


Figure 8 FTIR spectra of silica aerogel, control NRL, and NRL-silica aerogel film. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

of peak at 3362 cm^{-1} .³ The peaks at 1140 cm^{-1} and vibration at 822 cm^{-1} are evidents of Si—O bending interactions. This confirms the presence of silica aerogel in the NR host and existence of the interaction between NR and silica aerogel phases. However, the interaction is rather less significant as shown by the weaker peaks. This means that interaction between the NR macromolecular chains and silica aerogel is weak since polar silica aerogel is distributed in nonpolar hydrocarbon rubber.

Thermal properties

Thermogravimetric Analyser

Figure 9 shows the weight loss by thermal degradation of different filler NRL films. There is only one obvious thermal decomposition step in NR molecular chains which is thermal scissions of C—C chain bonds.

Data in Table III shows that weight losses occur at 300, 350, and 400°C. The results suggest that addition of silica aerogel into the rubber matrix increase the surface contact area between the silica aerogel particles and the rubber matrix, which consequently increase the absorption of heat energy during decomposition.²⁰

Thermal stability of prepared NRL thin film is dominated by the distribution of silica aerogel nanoparticles. Homogeneous distribution of nanoparticles

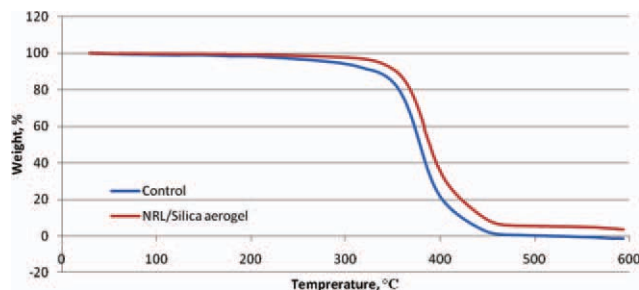


Figure 9 TG curves for NRL thin films in nitrogen. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

TABLE III
Weight Loss of NRL Films

Type of film	Weight loss at 300°C (%)	Weight loss at 350°C (%)	Weight loss at 400°C (%)	Total weight loss (%)
Control	5.71	14.29	77.86	97.86
NRL/silica aerogel	2.85	7.86	63.75	74.46

in NR matrix is achieved when the silica aerogel content is less than 4 phr. Strong interaction between silica aerogel and NRL molecular chains is promoted by various effects such as branching, size, nucleation, and surface properties.³ As a result, diffusion of decomposed products from bulk polymer to gas phase is slowed down. Consequently, improvement in thermal aging resistance of silica aerogel filled-NRL film is more pronounced compared to the pure NRL film. Furthermore, possible migration of inorganic particles to the surface of the films at elevated temperatures because of its relatively low surface potential energy is another reason for the improved aging resistance of silica aerogel filled NRL.²¹ This migration results in the formation of a NR/silica aerogel char, which acts as a heating barrier to protect the NR. Similar result was performed, where a clay/polymer char greatly enhances the thermal resistance of the host polymers.²²

Glass transition temperature (T_g)

The glass transition temperature (T_g) of a polymer is an important indicator of its thermal properties. The thermal behavior of NRL thin films with different types of fillers were analyzed by DSC. Figure 10 shows the DSC thermograms of NRL thin films with different filler types. The T_g was recorded as the half height of the corresponding heat capacity jump. As can be seen, the T_g of the composites was affected by addition of filler. The T_g of pure NRL and silica aerogel filled NRL thin films are 59.44 and -63.33°C , respectively.

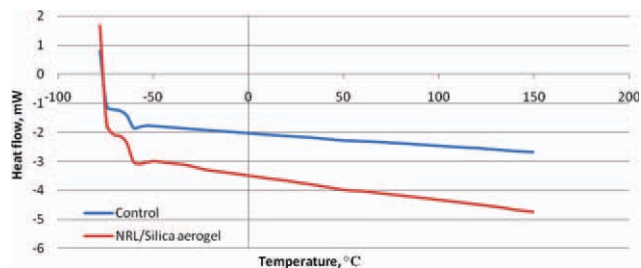


Figure 10 Glass transition temperature of different types of filler of NRL thin films measured from DSC. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

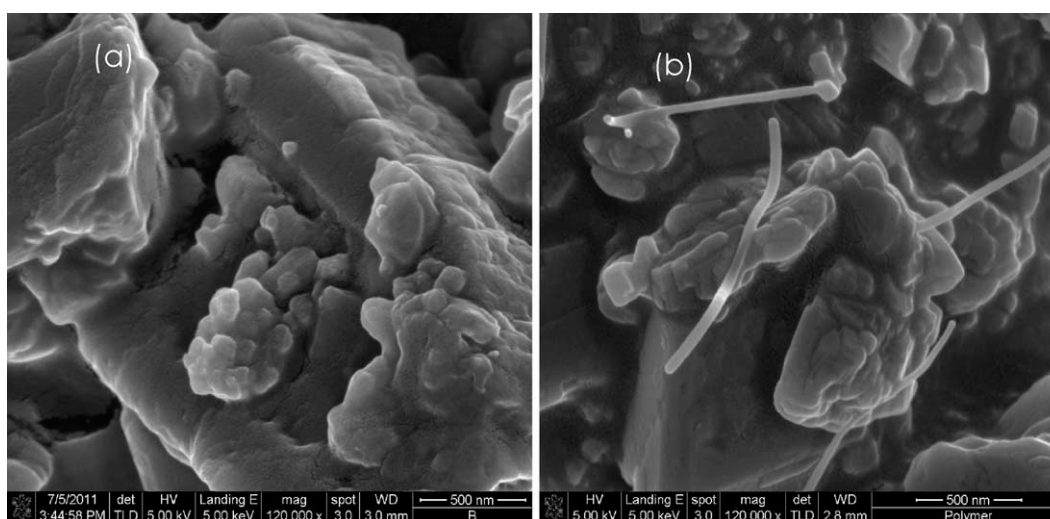


Figure 11 SEM micrographs of NRL (a) blank (b) silica aerogel 4 phr loading amount with leaching effect.

The DSC thermogram clearly indicates the existence of restricted segmental motion and NRL-silica aerogel interactions. This enables the polymer chains to absorb more energy in the form of kinetic energy. At the transition, the absorption of heat by the sample is increased, i.e., less heat is reflected from the surface. In this case, the increased absorption of heat by the sample was observed as a slower increase in the probe temperature.¹² Obviously, the addition of silica aerogel increases the glass transition temperature of NRL films, suggesting that silica aerogel plays a role in inhibiting segmental motion of NRL. Eisenberg's model explains that the interaction of polymer chains with silica reduces the mobility of polymer chains and leads to formation of immobilized and restricted mobility regions around the filler particles.^{12,13,17}

Differential scanning calorimetry (DSC) provides further information concerning intercalation. Increase in interactions between intercalated chains of polymer with the host species greatly reduces their rotational and translational mobility. The situation is similar to that in a reticulated polymer, where restrictions on its mobility increase its glass transition temperature (T_g). A similar increase is anticipated to occur in a nanocomposite due to elevation of the energy threshold needed for the transition.²³ Considerable increase in T_g is an important property of these materials which enables them to be resistant at higher temperatures compared with the original polymer and thus extends their fields of application.

Morphology of NRL films

When the surface area of filler is large, the filler is distributed homogeneously throughout the NR matrixes. On the other hand, filler with low surface

area tends to form aggregates. This is demonstrated in Figure 11(a,b), which shows the morphology of blank and silica aerogel filled NRL films, respectively. The SEM images magnified to 500 nm of both blank and filled NR films show similar morphology, with the NR matrix exists as the bulk phase. The surface of the filled NR film in Figure 11(b) appears rougher due to homogeneously embedded nanostructured silica aerogel particles forming the core-shell structure as shown by the TEM image in Figure 3. The absence of obvious phase separation indicates good miscibility between NR and silica aerogel.

CONCLUSIONS

Incorporation of inorganic silica aerogel nanoparticles into NRL matrix leads to an improvement in the mechanical properties of the host elastomer. Test results show that NRL films loaded with 4 phr of silica aerogel gives the optimum tensile, tear strength, and elongation at break. The glass transition temperature of the NRL-silica aerogel films is elevated by 3.78°C from the blank. Meanwhile, the mechanical properties of the NRL-silica aerogel are increased as were the tensile, tear strength, and modulus. Meanwhile, the mechanical properties of the NRL-silica aerogel were increased as were the tensile, tear strength, and modulus. All of these results indicate that silica aerogel is a good nanomaterial for use as an NRL film-reinforcing agent or filler and may improve the mechanical and thermal properties of NRL.

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