Preparation of Polydimethylsiloxane-Coated α-Alumina Fillers with Cold Plasma for Elastomer Thermal Interface Materials

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ABSTRACT: A novel method for the organic modification of a ceramic thermal conductive filler (α-alumina) with cold plasma was developed for the preparation of elastomer thermal interface materials with high thermal conductivities and low moduli. The α-alumina fillers were first coated with low-molecular-weight polydimethylsiloxane (PDMS) by solution dispersion and then treated in argon plasma for different time. The modified α-alumina fillers were characterized with high-resolution transmission electron microscopy, thermogravimetric analysis, Fourier transform infrared spectroscopy, and X-ray photoelectron spectroscopy. The results revealed that a thin PDMS film with several nanometers thick was tightly coated on the surface of the alumina filler after plasma treatment, and this thin film could not be removed by 48 h of Soxhlet extraction with *n*-hexane at 120°C. Plasma modification of the alumina could dramatically weaken the strength of the filler–filler networks and, thus, remarkably reduce the modulus of the alumina-filled silicone rubber composites but did not affect the thermal conductivity of the composites. © 2011 Wiley Periodicals, Inc. J Appl Polym Sci 123: 2875–2882, 2012

Key words: cold plasma; elastomers; fillers; surface modification; thermal properties

INTRODUCTION

With the continually diminishing size of electronic devices, heat dissipation has become one of the most critical problems that limit improvement in performance and reliability. It is highly desirable to design and fabricate thermal interface materials (TIMs) to improve heat transfer from the heat source to heat sink by a reduction of the thermal contact resistance caused by the air pockets between two contacting surfaces.[1,2] Elastomeric thermal padding (i.e., elastomer TIM) is one of most commonly used TIMs for its easy assembly, high reusability, and good damping effect.[2] Not only a high thermal conductivity and electric insulation but also a low modulus and high compliance are required for this material to provide a good thermal interface for uneven and rough surface topography. Elastomer TIMs are typically made up of a silicone matrix reinforced with highly thermally conductive but electrically insulating ceramic fillers, such as alumina, aluminum nitride, boron nitride, or silicon carbide.[3] To satisfy the requirement of high thermal conductivity, the loading of the thermal conductive filler is always very high, usually over 250 wt parts/100 wt parts silicone matrix. Obviously, such high filler contents must result in strong filler–filler networks in the composite and increase the modulus and hardness substantially.[4,5] The current solution to this problem is the organic modification of the filler surface with the use of coupling agents, which can increase the affinity between the filler and polymer and lower filler–filler interaction. α-Alumina has a high thermal conductivity and low cost;[6] this makes it a desirable conductive filler for elastomer TIMs. However, α-alumina is hard to sufficiently organically modify with traditional surface treatment approaches for satisfying the requirement of the material because of its low surface activity.

Surface modification by cold plasma was first developed in the 1960s and has been widely applied in the metallurgical, chemical, electronics, medicine, and environmental protection fields.[7–11] Cold plasma has many advantages for surface modification applications. For example, it has a high efficiency. The treatment will only change the properties of the material surface down to a depth of several to several 10s of nanometers rather than the bulk properties. For this reason, plasma treatment is considered a new type of surface modification method after conventional modification by coupling agents. Some researchers have studied the organic modification of inorganic fillers and polymers with plasma activity and polymerization.[12–16] So far, only a few investigations on plasma-modified alumina particles have been reported. For example, Shi et al.[17] grafted pyrrole onto the surface of alumina using plasma polymerization. In this study, we developed a new method for modifying alumina with a cold plasma process. The modified alumina was used for the preparation of high-performance elastomer TIMs with a high thermal conductivity, conformability, and electric insulation. In this method, low-molecular-weight, vinyl-terminated polydimethylsiloxane [PDMS; a liquid silicone rubber (LSR)] was used as the modifying agent, with the expectation that it would have good affinity to the PDMS matrix [i.e., silicone rubber (SiR)]. The state of the LSR coating on the modified alumina fillers was evaluated with high-resolution transmission electron microscopy (HRTEM), thermogravimetric analysis (TGA), Fourier transform infrared (FTIR) spectroscopy, and X-ray photoelectron spectroscopy (XPS). The amounts of LSR coating on alumina fillers under different plasma treatment time were estimated by the use of TGA and FTIR. The effects of the plasma treatment time on the amount of LSR coating and the modulus and thermal properties of the SiR composites filled with the LSR-coated alumina were studied.

EXPERIMENTAL

Materials

Methyl vinyl SiR (110-2 grade) and methyl silicone oil (201 grade) were purchased from Chenguang Research Institute and Chemical Industry (ChengDu, China). The vinyl-terminated, low-molecular-weight polydimethylsiloxane (i.e., LSR), with an average molecular weight of 120,000, was kindly provided by Nantong Silicone R&P Material Co., Ltd. (Nantong, China). The α-alumina filler (LM2-1014-E, purity $\approx$ 99.9%) was bought from Dalian Luming Nano-Material Co. (Dalian, China). All of these raw materials were dried at 160°C for 4 h before use to remove trace quantities of adsorption water and low-molecular volatile substances. Hexane (analytically pure) was purchased from Beijing Chemical Plant, (Beijing, China). Argon gas (purity > 99.5%) was provided by Beijing Ouya Gas Preparation Co. (Beijing, China).

Modification of the alumina with cold plasma

A predetermined amount of LSR (weight ratio of LSR to α-alumina = 3 : 100) was dispersed in hexane. Alumina fillers were added to the solution under stirring for a few minutes. The solution was first put into a vacuum oven at 80°C to remove most of the hexane and then completely dried at 160°C for 2 h. After the hexane disappeared, the powder was charged into a homemade plasma apparatus, similar to that reported in ref. [18]. Before the plasma reaction was activated, the pressure in the reactor was reduced to 25 Pa. Then argon gas was injected into the reactor, and the vacuum pressure of the plasma reactor was controlled at about 20 Pa with the help of throttle valves. The electrical power of the plasma was supplied by a radio-frequency power generator operating at 13.56 MHz and 90 W. The plasma treatment time ranged from 1 to 3 h in increments of 0.5 h.

In this article, we use L-alumina and LPx–alumina to stand for LSR-coated alumina and LSR-coated alumina after different plasma treatment time, where x is a variable number representing the plasma treatment time. For example, LP2–alumina stands for LSR-coated alumina after 2 h of plasma treatment. Some L–alumina and LPx–alumina fillers were subjected to 48 h of Soxhlet extraction with hexane at 120°C to remove the LSRs that were not tightly coated on the alumina. L–E-alumina and LPx–E-alumina refer to the alumina fillers subjected to the extraction. For example, LP2–E-alumina stands for LSR-coated alumina after 2 h of plasma treatment and subjected to 48 h of Soxhlet extraction with hexane at 120°C.

Preparation of the SiR/alumina composites

The filler (untreated alumina, L–alumina, or LPx–alumina), methyl silicone oil, and vulcanizing ingredients were mixed into the SiR matrix on a 6-in. two-roll mill φ 160 × 320, Zhanjiang machinery factory, Guangdong, China according to the recipe listed in Table I. The compound was vulcanized in a standard mold at a pressure of about 20 MPa and a temperature of 170°C for 20 min. The waferlike vulcanized sample, with a diameter of 60 mm and a thickness of 6 mm, was used for thermal conductivity measurements.

Characterization and measurements

HRTEM images were taken with a JEM-3010 transmission electron microscope (HRTEM; JEM-3010, Japan

TABLE I
Formulation of the SiR/Alumina Composites

Ingredient	Content (wt parts)
Methyl vinyl silicone rubber	80
Methyl silicone oil	20
LSR-coated alumina/untreated alumina	300
Triallyl isocyanurate	0.4
2,5-Dimethyl-2,5-bis(*tert*-butylperoxy) hexane	0.1

Electronics Co., Japan) at an accelerating voltage of 200 kV. A thermogravimetric analyzer (Star^e System TGA/DSC1, Mettler-Toledo, Switzerland) was used to estimate the quantity of LSR on the alumina particle surface. TGA was operated from room temperature to 800°C in nitrogen flow with a heating rate of 20°C/min. The FTIR absorption spectra of all of the samples were acquired with a Tensor 27 FTIR spectrometer (Bruker Optics, Germany) in the wave-number range 4000–400 cm^{-1} (KBr pellet technique) at a resolution of 4 cm^{-1}. XPS analysis was carried out on an ESCA LAB 250 (Thermo Fisher Scientific Corp., USA) with an Al Kα X-ray source (1486.6-eV photons). The incidence angle was 45°, and the corresponding escape depth for the photoelectrons was 10 nm. The strain amplitude dependence of the dynamic storage modulus (G') of the SiR/alumina compound was measured with a rubber process analyzer (RPA2000, Alpha Technologies, USA). A strain sweep from 0.55 to 325% was performed at 60°C and 1 Hz. The fractural surface of the SiR/Al$_2$O$_3$ composite was observed with scanning electron microscopy (SEM; S-4700, Hitachi, Japan). Thermal conductivity was measured with an HC-110 thermal conductivity tester (Laser Co., USA) according to ASTM C518-98.

RESULTS AND DISCUSSION

Characterization of the cold-plasma-modified α-Al$_2$O$_3$

Figure 1(a) displays the HRTEM image of the untreated alumina particles. The image clearly shows the naked particle surface of alumina, and the particle size was about 200 nm. Figure 1(b) shows the HRTEM image of LP2–E-alumina and demonstrates an ultrathin coating of 2–3 nm thick on the surface of the alumina particles. The latter image provides direct evidence for that LSR was tightly coated on the alumina surface by plasma treatment.

TGA was used to estimate the mass of the LSR coating on the surface of the alumina particles. Figure 2 demonstrates the TGA profiles of L–alumina. It could be observed that LSR began to decompose at 410°C, and the decomposition ended at 590°C. From the weight loss of this decomposition, the LSR content of L–alumina was estimated to be 2.88%; this corresponds to a LSR/alumina ratio of about 2.965/100. Because the amounts of LSR and alumina actually used were in the ratio 3/100, we concluded that the majority of LSR was successfully coated on the alumina particle surface by solution dispersion.

Figure 3 displays the FTIR spectrum of L–alumina and LP*x*–alumina before and after 48 h of Soxhlet extraction with *n*-hexane at 120°C. Besides several strong absorption peaks in the range 480–1000 cm^{-1}, corresponding to alumina, and the absorption peaks at 3450 and 1650 cm^{-1}, corresponding to the stretching and deforming vibration modes, respectively, of the H—O—H bonds of the adsorbed water, the small absorbances ranging from 1000 to 1150 cm^{-1}, assigned to the Si—O—Si stretching mode,[19,20] and the small
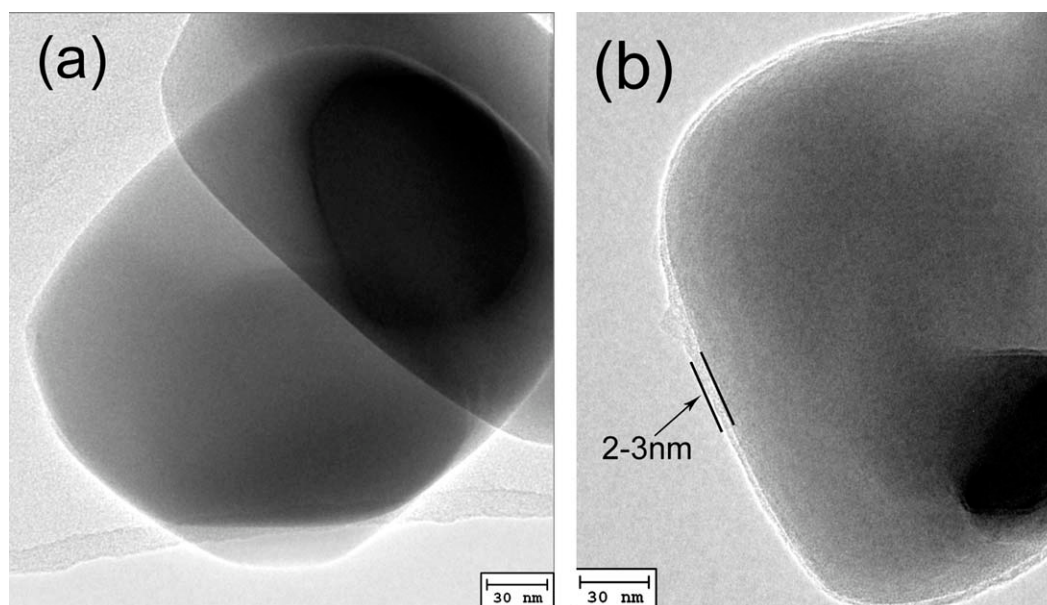

Figure 1 HRTEM images of (a) untreated alumina and (b) LP2–E-alumina (LSR-coated, 2-h plasma-treated alumina after 48 h of extraction).
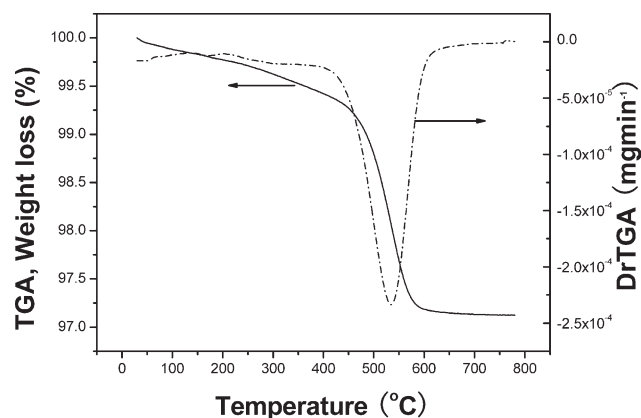
Figure 2 TGA and derivative TGA (DrTGA) profiles for LSR-coated alumina (L–alumina).

absorbance at about 1260 cm^{-1}, assigned to the C—Si stretching mode,[20] were obvious; this indicated the presence of SiR. Comparing the insets in Figure 3(a,b), we can see that LPx–alumina and LPx–E-alumina showed different trends in the absorbance of the Si—O—Si bands with plasma treatment time. Before extraction [see Fig. 3(a)], the absorbance of the Si—O—Si bands (1000–1150 cm^{-1}) basically decreased with increasing plasma treatment time; this indicated a reduction in the entire amount of LSR coating. This phenomenon could be explained by the degradation of the LSR coating caused by the plasma initiation gas (Ar).[21] The absorbance peaks associated with LSR were weaker after extraction than those before extraction. It is believed that 48 h of Soxhlet extraction with hexane at 120°C was sufficient to remove any LSR physically adsorbed on the alumina surface. So we inferred from the results shown in Figure 3(b) that some LSR was tightly coated on the alumina surface by cold plasma treatment. We speculated that a crosslinked network of LSR might have been induced by plasma treatment and resulted in a tight coating of LSR that could not be removed from the alumina surface by extraction. From the inset in Figure 3(b), it can also be seen that the absorbance intensity of the Si—O—Si bands increased with increasing plasma treatment time until a maximum value was reached at 2 h (i.e., LP2–E-alumina). The absorbance intensity dropped after 2 h; this suggested that 2 h was the optimal plasma treatment time for the formation of tightly coated LSR in this work.

As shown in Figure 3, all of the FTIR spectra were normalized with the absorbance of the alumina bands (ranging from 480 to 1000 cm^{-1}) as the intrareference. Therefore, the content of LSR coating [i.e., the coating rate (CR)] for modified alumina could be determined quantitatively according to the normalized absorbance intensity of the Si—O—Si stretching bands and the LSR content of L–alumina (i.e., 2.88%) derived from

TGA (see Fig. 2). According to FTIR results, as shown in Figure 3, there were two types of LSR coating on the alumina surface after plasma treatment: tightly coated and physically adsorbed LSRs. Hence, we defined two types of CR, CR(entire)[22] and CR(tight), which could be estimated by following equations:

$$CR(entire)(\%) = \frac{2.965 \times \frac{A}{A_0}}{100 + 2.965 \times \frac{A}{A_0}} \times 100 \quad (1)$$

$$CR(tight)(\%) = \frac{2.965 \times \frac{A'}{A_0}}{100 + 2.965 \times \frac{A'}{A_0}} \times 100 \quad (2)$$

where CR(entire) and CR(tight) represent the LSR contents of LPx–alumina and LPx–E-alumina, respectively, and A, A', and A_0 represent the normalized absorbances of the Si—O—Si bands of LPx–alumina, LPx–E-alumina, and L–alumina, respectively.
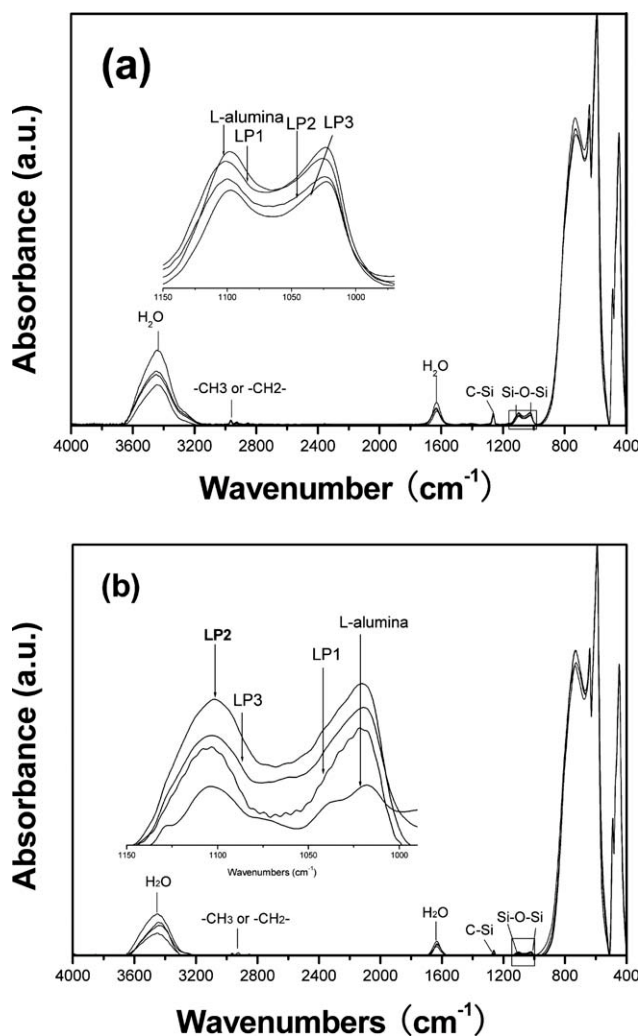

Figure 3 FTIR spectra for LSR-coated alumina with different plasma treatment time: (a) before extraction and (b) after 48 h of extraction. All spectra were normalized by the absorbance of alumina around 480–1000 cm^{-1}.
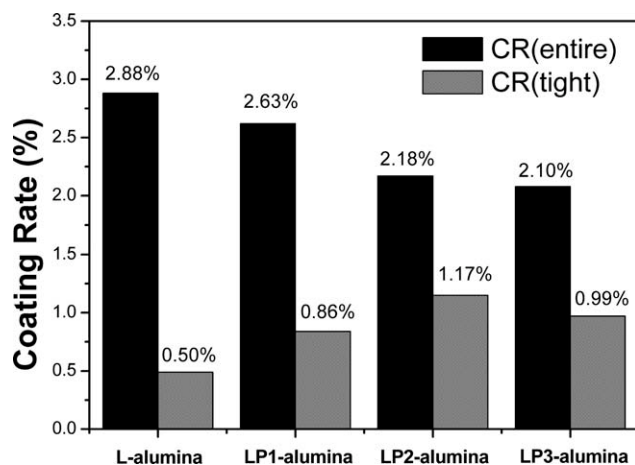
Figure 4 CR for different treated alumina fillers.

Figure 4 shows the results of CR(entire) and CR(tight) for different alumina samples. It can be seen that CR(entire) decreased from 2.88 to 2.10% with increasing plasma treatment time. On the other hand, CR(tight) increased first, reached a peak value of 1.17% at the plasma treatment time of 2 h, and then decreased. For the L–alumina sample without plasma treatment, there was still a little LSR [CR(tight) ≈ 0.50%] remaining after the extraction; this was probably a result of the crosslinking reactions of LSR during the 2 h of drying at 160°C. From CR(tight), the thickness of the LSR film tightly coated onto the alumina surface could also be estimated with the following equations:

$$\frac{m_1}{m_2} = \frac{\rho_1 \times V_1}{\rho_2 \times V_2} = \frac{\rho_1 \times \frac{4}{3}\pi R_A^3}{\rho_2 \times \frac{4}{3}\pi (R_{LA}^3 - R_A^3)} \quad (3)$$

$$\frac{m_1}{m_2} = \frac{[100 - CR(tight)]}{CR(tight)} \quad (4)$$

$$LSR \text{ film thickness } = R_{LA} - R_A \quad (5)$$

where m, ρ, and V represent the mass, density, and volume, respectively; the subscripts 1 and 2 stand for alumina and LSR, respectively; and R_A and R_{LA} represent the radii of pristine alumina and LSR–tightly coated alumina particles, respectively. From Figure 1, R_A was estimated to be about 100 nm. Using LP2–alumina as an example [CR(tight) = 1.17%], we obtained a calculated thickness of about 2.38 nm for the tightly coated LSR film; this was in good agreement with the HRTEM result shown in Figure 1(b). This agreement confirmed the validity of the CR calculations from the TGA and FTIR results.

The wide-scan XPS spectra of untreated alumina and LSR-coated alumina after different times of plasma treatment (i.e., LPx–alumina samples) are shown in Figure 5. In the spectrum of the untreated alumina, there were Al 2p and Al 2s core-level peaks

at 74.8 and 118.8 eV[23] associated with alumina. It should be pointed out that the positions of the Al 2p and Al 2s core-level peaks for the LPx–alumina samples were almost same as those for untreated alumina. This suggested that the plasma treatment did not change the microstructure of the alumina fillers and allowed the high thermal conductivity of the alumina filler to remain. Moreover, the Si 2p and Si 2s core-level peaks appeared at 102.2 and 152.9 eV; this indicated the presence of coated LSR, and their intensities decreased with increasing plasma treatment time. The wide-scan XPS spectra of LSR-coated alumina after different plasma treatment time and after 48 h of extraction (i.e., LPx–E-alumina samples) are shown in Figure 6. In XPS spectra of all of the LPx–E-alumina, the Si 2p and Si 2s core-level peaks still existed at about 102.2 and 152.9 eV, respectively. This indicated the presence of tightly coated LSR that could not be removed from the alumina surface by extraction. According to the XPS spectra shown in Figures 5 and 6, the atom contents of Si 2p in the surface (measuring depth ≈ 10 nm) of differently treated alumina were determined, as summarized in Table II. The following phenomena were found:

1. The content of Si 2p for LPx–alumina decreased with increasing plasma treatment time. This indicated that CR(entire) decreased with increasing treatment time.
2. The content of Si 2p for the LPx–alumina samples was larger than that for the corresponding LPx–E-alumina.
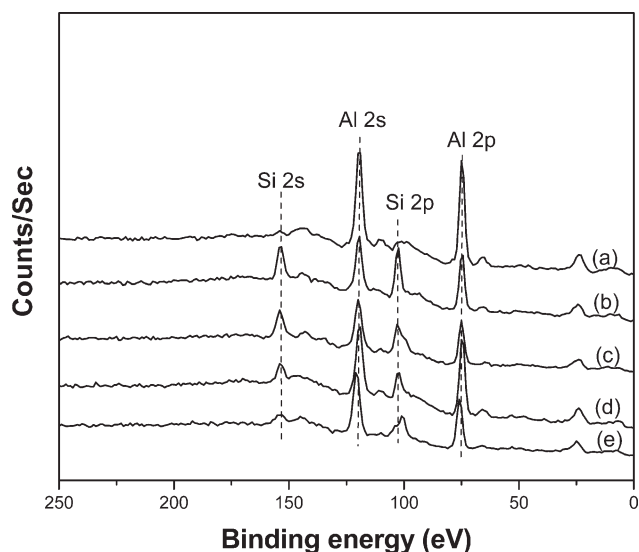

Figure 5 XPS wide-scan spectra of (a) untreated alumina, (b) LP0–alumina (LSR-coated alumina without plasma treatment), (c) LP1–alumina (LSR-coated, 1-h plasma-treated alumina), (d) LP2–alumina (LSR-coated, 2-h plasma-treated alumina), and (e) LP3–alumina (LSR-coated, 3-h plasma-treated alumina). The spectra were vertically shifted for clarity.
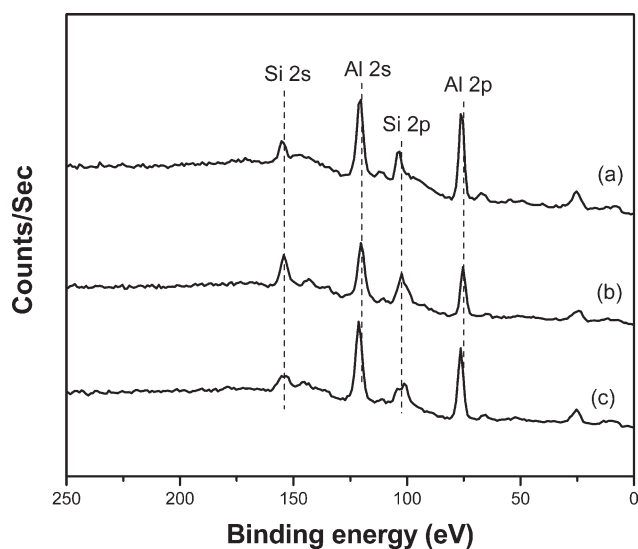
Figure 6 XPS wide-scan spectra of (a) LP1–E-alumina (LSR-coated, 1-h plasma-treated alumina after 48 h of extraction), (b) LP2–E-alumina (LSR-coated, 2-h plasma-treated alumina after 48 h of extraction), and (c) LP3–E-alumina (LSR-coated, 3-h plasma-treated alumina after 48 h of extraction). The spectra were vertically shifted for clarity.

3. Among the LP*x*–alumina samples, the Si 2p content of the LP2–E-alumina was the highest. This indicated that 2 h of plasma treatment was the optimum condition for obtaining the highest CR(tight).

These XPS results further verified the changing trends of CR(entire) and CR(tight) with plasma treatment time derived from TGA and FTIR experiments.

Influence of plasma modification on the filler–filler networks and the thermal conductivity of the SiR/alumina composites

The strain–amplitude dependence of G' of filled rubber was brought into clear focus by the work of

TABLE II
Si 2p Atom Contents of Different Alumina Samples Derived from XPS

Sample	Si 2p content (atom %)[a]
LP0–alumina[b]	20.57
LP1–alumina	14.69
LP2–alumina	12.35
LP3–alumina	11.40
LP1–E-alumina	7.54
LP2–E-alumina	8.78
LP3–E-alumina	7.94

[a] The starting and ending binding energies for Si 2p content calculation are 92.53 and 105.20 eV, respectively.
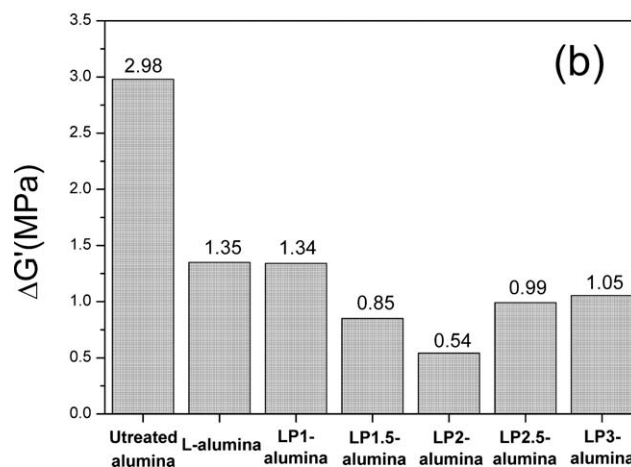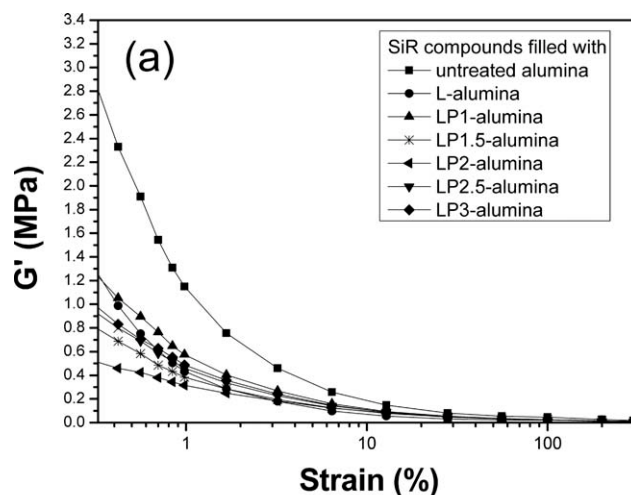[b] LP0–alumina stands for LSR-coated alumina without plasma treatment.

Figure 7 Rubber process analysis results of different SiR/alumina compounds: (a) G' as a function of strain and (b) $\Delta G'$.

Payne in the 1960s and is now often referred to as the *Payne effect*.[24] It is used to qualitatively characterize filler–filler networks. For filler–rubber systems, the higher the dynamic storage modulus is at initial strain (G'_0) and the quicker the G' attenuation is with strain, the stronger the filler–filler networks and Payne effect will be. $\Delta G'$, the difference between G'_0 and the ultimate dynamic storage modulus (G'_∞) at high strain, is considered a qualitative reflection of the filler–filler network strength.[25] The Payne effects of the SiR compounds filled with different alumina were examined with rubber process analysis, the results of which are shown in Figure 7. From Figure 7(a), it can be seen that the compound with untreated alumina as a filler exhibited the strongest Payne effect and had the highest G'_0 of 2.98 MPa. The use of LSR-coated alumina (i.e., L–alumina) as a filler significantly reduced the Payne effect of the filled SiR compound. Plasma treatment (i.e., with LP*x*–alumina as the filler) further reduced the Payne effect, and the degree of reduction depended on the plasma treatment time. As shown in Figure 7(b),

TABLE III
Comparison of the Thermal Conductivities for Different SiR/Alumina Composites

Sample	Untreated alumina	L–alumina	LP2–alumina	Comparison 1 (untreated alumina)[a]	Comparison 2 (untreated alumina)[b]
Apparent loading of alumina (wt parts)	300	300	300	291.36	293.49
Actual weight concentration of alumina (wt %)	74.91	72.75	73.28	72.75	73.28
Thermal conductivity (W m^{-1} k^{-1})	1.34	1.22	1.23	1.22	1.24

[a] We added 8.64 (300 − 291.36) weight parts LSR to the compound to replace the alumina.
[b] We added 6.51 (300 − 293.49) weight parts LSR to the compound to replace the alumina.

the compound with LP2–alumina as a filler had the lowest $\Delta G'$ of 0.54 MPa, which was about 18% that of the compound filled with untreated alumina.

The SEM images of the fracture surfaces SiR filled with 300-phr untreated alumina and 300-phr LP2–alumina (i.e., LSR-coated alumina with 2 h of plasma treatment) are displayed in Figure 7(a,b), respectively. It can be seen that the dispersion and packing state of pristine alumina and LP2–alumina in the SiR matrix were almost same under the condition of high filling. This indicated that the packing and dispersion states were not the main reason for the dramatic reduction of the Payne effect of the SiR compounds filled with LPx–alumina.

Recall that CR(entire) for treated alumina followed the following order: LP2–alumina > LP3–alumina > LP1–alumina > L–alumina. Nevertheless, the reverse order was true for $\Delta G'$ of the SiR/alumina compound. These results suggest that the higher the amount of LSR tightly coated onto the alumina particles was, the weaker the filler–filler interactions were. Figure 1 shows that plasma treatment could produce a nano-LSR film tightly coated onto the alumina particle surface. This LSR nanofilm not only improved the interfacial affinity between the alumina filler and the matrix SiR but also prevented the alumina particles from directly contacting each other. As a result, the strength of the filler–filler networks of the SiR/alumina compound was dramatically reduced, and a highly filled SiR/alumina compound having a low modulus (i.e., ultrasoft) could be obtained.

Thermal conductivity is another primary performance requirement, besides compatibility, for elastomer TIM. Table III shows that the composite with untreated alumina as a filler had a thermal conductivity of 1.34 W m^{-1} k^{-1}. The thermal conductivities of the SiR/L–alumina and SiR/LP2–alumina composites were 1.22 and 1.23 W m^{-1} k^{-1}, respectively. It appeared that the LSR coating of alumina would lower the thermal conductivity of these SiR/alumina composites. However, the actual alumina content in these two SiR composites was lower than that in the SiR/untreated alumina composites, despite the same apparent loading of alumina of 300 parts by weight.

A supplement experiment was performed involving two samples filled with pristine alumina with the same actual alumina contents as those used in SiR/L–alumina and SiR/LP2–alumina. Table III shows that SiR/untreated alumina, SiR/L–alumina, and SiR/LP2–alumina had about the same thermal conductivity at the same actual alumina content.

At present, it is well accepted that thermal conductive fillers need to touch one another to form a continuous heat conduction path in the filled polymer
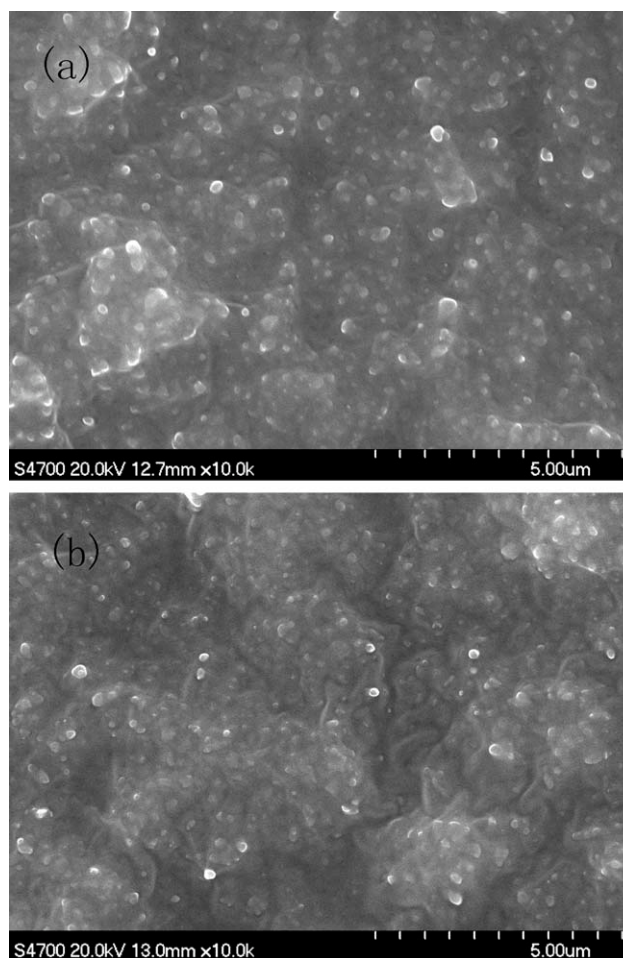

Figure 8 SEM photographs of the fracture surfaces of the SiR/Al$_2$O$_3$ (100/300) composites filled with (a) untreated alumina and (b) LP2–alumina (LSR-coated, 2-h plasma-treated alumina).

composites to achieve a high thermal conductivity.[26] The formation of a heat path is related to the distribution and packing state of the thermal fillers in the polymer matrix and to the surface roughness. SEM observation of the SiR/Al$_2$O$_3$ composites (Fig. 8) demonstrated that the dispersion and packing state of the untreated alumina and LP2–alumina in the SiR matrix was similar when the filler loading was high (i.e., 300 phr). Furthermore, HRTEM images of the untreated alumina and LP2–E-alumina shown in Figure 1 revealed that the plasma treatment did not change the surface roughness of the filler. Therefore, our results imply that the LSR thin film (i.e., 2–3 nm thick) coated on the alumina surface did not obstruct the formation of the heat-flow path in the SiR/alumina composite. Some further works to verify this assumption are ongoing; they will be reported in the future.

CONCLUSIONS

Alumina filler was first coated by LSR by solution dispersion and was subsequently treated by argon cold plasma. This modification method produced a nano-film of LSR tightly coated onto the surface of alumina particles. The amount of tightly coated LSR [i.e., CR(tight)] varied with the plasma treatment time, and the optimal coating time was found to be 2 h, at which time the resulting CR(tight) was 1.17% and the thickness of the tightly coated LSR film was 2–3 nm.

The modification of alumina with LSR coating and plasma treatment dramatically weakened the strength of the filler–filler networks and lowered the modulus of the SiR/alumina composite. The higher the amount of LSR coated onto the alumina surface was, the lower the modulus of the SiR/alumina composite was. According to XPS analysis, plasma treatment did not change the structure of the α–alumina filler. SiR filled with LSR-coated alumina fillers retained the high thermal conductivity of SiR filled with untreated alumina. Therefore, the plasma modification for ceramic thermal conductive fillers proposed in this study should be very favorable to the preparation of ultrasoft elastomer TIMs.

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