Efficiently Enhancing Low-dielectric Properties and Chemical Resistances of Addition-cure Liquid Silicone Rubber by Filling With Silicone-modified Eucommia Gum Nanofiller

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Abstract

It is of great interest and remains a challenge to simultaneously improve the low-dielectric properties and chemical resistances of addition-cure liquid silicone rubber (ALSR). In this work, we proposed an efficient approach to address this issue by filling silicone-modified eucommia gum nanofiller (SEUG) into ALSR. By adding 5 wt% SEUG, the dielectric constant of the SEUG/ALSR composite rubber was significantly lower than that of neat ALSR both at 10^2 Hz, 10^3 Hz and 10^4 Hz. Simultaneously, the SEUG/ALSR composite rubber exhibited better mechanical and insulation properties than the neat ALSR after HCl, NaCl, and oil resistance tests. Our findings demonstrated the great potential for fabricating silicone rubber with low-dielectric properties and erosion resistance by the utilization natural biomass rubber material, endowing it with excellent mechanical performance and degradability.

Introduction

Addition-cure liquid silicone rubber (ALSR) is a packaging material that has been widely application in electronics, electrical power, and electrical fields [1-5] because of its excellent high- and low-temperature resistance, good elastic properties, and lack of by-product residues or volatiles. However, practical application of pure ALSR is severely hindered by its poor mechanical properties [6,7] and poor erosion resistance [8-14]. Packaging material failure due to corrosion often causes great harm to electronic components, which significantly threatens the security of electrical systems. Therefore, it is imperative to develop an effective strategy to enhance the mechanical properties and erosion resistance of ALSR. To overcome these problems, an effective method is to disperse nanoparticles into the ALSR, such as clay [15], TiO_2 [16-19], SiO_2 [20-23], graphene [24-27], carbon nanotubes [28,29], boron nitride (BN) [30,31], and aluminum nitride (AlN) [32]. However, due to the high permittivities and poor compatibility of these materials with ALSR, the dielectric constant of the ALSR significantly increases [33]. However, ALSR is widely used as an insulating packing material for electronic devices, such as central processing units and integrated circuit, and thus, a low dielectric constant and loss factor are required [34,35]. A low dielectric constant helps to decrease the time of signal propagation, while a low loss factor aids in decreasing the heat production. Such disadvantages limit the further applications of ALSR. Therefore, it is necessary to develop a new method to modify the ALSR for the improvement of the low dielectric constant and erosion resistance. Nevertheless, very few studies on ALSR have addressed this issue.

Eucommia ulmoides gum (EUG) is a special natural rubber obtained from eucommia ulmoides Oliv, which grows mainly in China. EUG exhibits many excellent properties due to its molecular structure containing long-chain trans-1,4-polyisoprene, such as strong insulation properties, a low dielectric constant, water resistance, chemical resistance, and a high toughness. Furthermore, the development of EUG is listed in the national key research program of China because EUG is renewable and degradable. Nevertheless, some drawbacks of pure EUG, such as its poor high-temperature resistance and elastic properties, limit its widespread application. Instead, EUG shows good corrosion protection properties as the nanofiller in composite materials and has shown great potential for application in anticorrosion materials. For example, Bo Chen et al. [36] added an epoxidized biomass eucommia gum into an epoxy

composite coating to obtain excellent anticorrosive performance. To the best of our knowledge, there have been few studies on EUG fillers in silicone rubber.

In this work, we selected silicone-modified eucommia gum nanofiller as the filler to systematically study its effects on the mechanical, insulation, dielectric, and chemical-resistance properties of ALSR. Through this study, we expect to contribute to the preparation of ALSR with extraordinary dielectric properties and chemical resistance. This work also provides a new and feasible strategy for manufacturing degradable ALSR.

Experimental

2.1 Materials

Eucommia ulmoides gum (EUG) was purchased from Xiangxi Laojiao Biological Co., Ltd. (China). Xylene and hydrogen peroxide (30% wt) were purchased from China National Pharmaceutical Group Co. The platinum catalyst (platinum content was 5000 ppm) and 1-ethynylcyclohexanol were supplied by Shin-Etsu Co., Ltd., Japan. Poly(hydromethylsiloxane)-1 (PHMS-1, 0.1 wt% hydride), Poly(hydromethylsiloxane)-2 (PHMS-2, 0.5 wt% hydride), and vinyl-terminated poly(dimethylsiloxane) (VPDMS, 0.28 wt% vinyl) were obtained from Elkem Silicones Co., Ltd., China.

2.2 Synthesis of silicone-modified eucommia gum nanofiller (SEUG)

Silicone-modified eucommia gum nanofiller (SEUG) was synthesized through our developed two-step hydrosilylation sol-gel reaction, which is shown in Fig. 1. In step one, the eucommia ulmoides was purified. The eucommia ulmoides shell (150 g) was cut into small particles, which were then transferred to a four-necked 5000-mL round-bottom flask equipped with a magnetic stirrer, and xylene (3500 g) was added. After this, the mixer was heated to 80°C, and a mixture of H₂O₂ (10 g) and pure water (150 g) was added dropwise under stirring. The mixture was stirred for 6 h at 80°C. Eucommia ulmoides (3538 g) was obtained after centrifugation to remove the impurities and water.

In step two, the hydrosilylation reaction occurred. The eucommia ulmoides after purification (3538 g) and the platinum catalyst (0.5 g) were added to a 5000-mL four-neck round bottom flask. The mixture was first stirred for 0.5 h at 60°C. Poly (hydromethylsiloxane) (PHMS-1 100g) was added dropwise to the flask. The mixture was stirred for 3 h at 60°C and then heated to 90°C for 3 h. Reduced-pressure distillation was appl

2.3 Preparation of the SEUG/ALSR composite rubbers

VPDMS (100 g) and SEUG (5, 10, or 15 g) were uniformly mixed in a high-speed disperser to obtain the nano-dispersion system. A suitable quantity of PHMS-2, 1-ethynylcyclohexanol, and Karstedt's catalyst was vigorously stirred using an electric mixer. The mixture was subsequently vulcanized at 120°C for 1 h, after which the ALSR samples were successfully prepared. The compositions of the ALSR samples are

listed in Table 1. The SEUG was uniformly dispersed in the ALSR matrix. To remove the xylene, and SEUG(135) was obtained. The synthesis scheme is shown in scheme. 1.

2.4 Characterization

2.4.1 Fourier-transform infrared spectroscopy

The liquid sample was first coated on the surface of the KBr slice and was then analyzed by Fourier-transform infrared spectroscopy (FT-IR, Tensor 27, Bruker Optics, Germany) from 4000 to 400 cm^{-1} with a resolution of 4 cm^{-1} and 16 total scans.

2.4.2 Surface morphology test

The fractured cross sections of the nanocomposites were characterized by scanning electron microscopy (SEM, Hitachi S-4700).

2.4.3 Mechanical properties test

The tensile strength and elongation at break of the ALSR specimens were measured using a universal testing machine (HD-WDW-100A, Xiamen HaiDa Inc., China) according to ISO 37-2005. The Shore A hardness of the samples was conducted with a Shore A durometer (LX-A, Shanghai Liuling Instruments Factory, China) in accordance with ISO 7619-1-2004.

2.4.4 Insulation performance test

The volume resistivities of the ALSR specimens were measured using a surface volume resistivity testing machine (ZST-121, ZhongHangShiDaixin Instrument Equipment Inc., China) according to ASTM D 257.

2.4.5 Dielectric performance test

The dielectric constant of the SEUG/ALSR composite rubber specimens were measured using a dielectric analyzer DEA-2970 coupled with a temperature controller 2000 (TA Instruments Inc., USA) using the compression-molded specimens with thicknesses of 0.3–0.6 mm. The measurements were performed at a frequency of 10^2 Hz, 10^3 Hz and 10^4 Hz.

2.4.6 Chemical resistance test

The chemical resistance of the ALSR was tested according to ISO 1817-2005. The effects of liquids (acid (HCl), base (NaOH), salt (NaCl), and aviation oil) on different ALSR samples were studied. In each case, five pre-weighed samples were dipped in the respective chemical reagents for 30 d. They were then removed, immediately washed with distilled water, and dried by pressing both sides of the samples with filter paper at room temperature. The mechanical properties and insulation properties of the ALSR samples were measured. The variation of the performance were calculated.

Results And Discussion

3.1 Characterization of SEUG

To address the issue of poor compatibility, the SEUG was synthesized through a hydrosilylation reaction, which is shown in Fig. 1. Fig. 2 shows the FT-IR spectra of the SEUG and EUG from which the functional groups were identified. The characteristic peaks at 1460 and 1380 cm^{-1} were attributed to the bending vibrations of the methyl groups, which indicated that EUG and SEUG both contained methyl groups. The peak at 1665 cm^{-1} was attributed to C=C stretching vibrations. The characteristic peak at 2869 cm^{-1} was assigned to the stretching vibrations of the CH in $\text{R}_2\text{C}=\text{CHR}$. The relative intensity of the C=C peak for the SEUG was significantly weakened compared to that of the EUG, suggesting that the C=C participated in the hydrosilylation reaction. However, the characteristic peak was still present, which indicated that C=C was present in the SEUG. Additionally, as for the SEUG, one new peak appeared at 2128 cm^{-1} , which was attributed to Si-H. The peaks at 1099 and 1024 cm^{-1} were characteristic of Si-O-Si groups. Thus, the silanization of EUG occurred through the hydrosilylation reaction.

3.2 Surface morphologies of SEUG/ALSR composite rubbers

To further confirm that SEUG was dispersed uniformly in the ALSR, observation of the morphologies of the SEUG/ALSR composite rubbers and neat ALSR were examined using SEM. The surface of the neat ALSR was smooth (Fig. 3, ALSR-1). Although uniform plications were evident on the surfaces of the composite rubbers containing 5 wt% SEUG (Fig. 3, ALSR-2) none evident independent particles was showed. This was evident where the dispersion of the SEUG nanofiller was sparse in the ALSR-2, which verified that most of the SEUG nanofiller was encapsulated in the ALSR.

The uniformity of the plications also indicated that the compatibility of the ALSR with the SEUG was significantly enhanced. However, when the doped contents of the SEUG further increased to 10 wt%, the surface of the SEUG/ALSR composite rubber (Fig. 3, ALSR-3) showed showed some evident particles compared to ALSR-2. The small particles were free macromolecular SEUG, and they could aggregate to form large particles. As shown in Fig. 3 (ALSR-4), upon further increasing the SEUG content to 15 wt%, the number of small particles was reduced, while the volumes of the large particles were larger than those of the ALSR-3. This verified that more particle aggregation occurred.

3.3 Mechanical properties of SEUG/ALSR composite rubbers

To understand the effect of the SEUG on the mechanical properties of the composite rubbers, the tensile strength, elongation at break, and Shore A hardness were determined. The tensile strength of the SEUG/ALSR composites as a function of the mass fraction of SEUG is shown in Fig. 4. In the case of the neat ALSR, the tensile strength value was about 1.35 MPa. For the composite rubbers with different doped contents of SEUG, the tensile strength increased first as the doped content of SEUG increased. The maximum reached 1.71 MPa when the content of SEUG was 5%. This can be explained as follows. On the one hand, SEUG had a larger relative molecular mass and longer molecular chains. Thus, it possessed

a higher mechanical strength than the ALSR, which led to increased tensile strengths of the SEUG/ALSR composite rubbers. On the other hand, the Si-H groups in the SEUG could react with the VPDMS through an addition reaction during the curing process, resulting in the formation of a closer interpenetrating network structure. The promotion of the tensile strength was the direct result of the richer crosslinking densities. However, the tensile strength decreased abruptly to 1.21 MPa after increasing the content of the SEUG nanoplate filler to 10 wt%. This indicated that with further addition of SEUG, the free SEUG could further form large particles due to agglomeration. The miscibility and compatibility of the SEUG with the ALSR could be reduced, thereby leading to a decreased mechanical strength of the SEUG/ALSR composite rubbers, which was confirmed by compatibility tests, as shown in Fig. 3.

The elongation at break and Shore A hardness values of the SEUG/ALSR composite rubbers are shown in Figs. 4 and 5, respectively. As the content of the SEUG nanoplate filler increased, the Shore hardness steadily decreased from 32 to 18. In contrast, the elongation at break of the ALSR strongly increased with the SEUG content. This indicated that the toughening of the SEUG/ALSR composite rubbers increased compared to that of the neat ALSR. This agreed with recently reported results [37,38].

3.4 Dielectric performance of SEUG/ALSR composite rubbers

The dielectric constants (ϵ_r) of the pure ALSR and SEUG/ALSR composites were measured at frequency of 10^2 Hz, 10^3 Hz and 10^4 Hz at room temperature, and the results are shown in Fig. 6. As the SEUG concentration increased, the dielectric constant first decreased and then increased. The possible mechanism through which the SEUG affected the low-dielectric properties of the ALSR is demonstrated in Fig. 7. Based on previous research, the relative dielectric constant of the filled ALSR was found to be governed by the mobility of the PDMS chains, the inherent permittivity of the nanofiller, and the polarization associated with the silicone and particles [39,40]. It is well-known that the SEUG had similar permittivities (2-3.2) to that of pure ALSR (2-3). Therefore, the influence of the SEUG nanofiller's inherent permittivity on the overall permittivities of the composites was negligible. As for the composite rubber with a SEUG doping content of 5 wt% (ALSR-2), the dielectric constant was lower than that of the neat ALSR both at 10^2 Hz, 10^3 Hz and 10^4 Hz. This suggested that the introduction of SEUG enhanced the low dielectric properties of the composite rubber compared to those of the neat ALSR. This phenomenon can be explained as follows. On the one hand, the SEUG/ALSR composite exhibited good dispersion of the SEUG particles. The SEUG particles were highly encapsulated by the ALSR matrix (Fig. 3, ALSR-2). No distinct interfacial polarization between the SEUG and ALSR phases was evident because of the hydrosilylation reaction during the surface modification of the SEUG nanoparticles. On the other hand, the Si-H groups in the SEUG could react with the VPDMS through an addition reaction during the curing process, resulting in the formation of closer interpenetrating network structures, which restricted the mobility of the polysiloxane chains in the composite rubbers. This restriction inhibited the electrical polarization because the material lost its freedom to relax under an applied voltage, and thus, the dielectric constant decreased. However, as the concentration of SEUG further increased, the dielectric constant of the composite rubbers exhibited a notable rise with increasing SEUG content, which was ascribed to the interfacial polarization (Maxwell–Wagner polarization) at the interface between the ALSR

or SEUG. As shown in Fig. 8, the small SEUG particles were created by the aggregation of free macromolecular SEUG, and the small SEUG particles further aggregated to form large SEUG particles. The particle agglomeration was confirmed by the SEM analysis. The presence of more interfacial polarization at the interface between the ALSR and SEUG particles resulted in a higher permittivity as the SEUG concentration increased.

3.5 Chemical resistance properties of SEUG/ALSR composite rubbers

To understand the effect of the SEUG on the chemical resistance properties of the composite rubbers, the tensile strength (mechanical properties) and volume resistivity (insulation performance) were measured after different chemical resistance tests. The effect of the SEUG on the chemical resistance properties of the SEUG/ALSR composite rubbers could be observed by the performance of tensile strength and volume resistivity. The performance after the HCl, NaCl, and oil resistance tests with different SEUG contents is shown in Figs. 8, 9, and 10, respectively.

It could be seen that the tensile strength and volume resistivity of the SEUG/ALSR composite rubbers after the HCl, NaCl, and oil resistance tests were similar. The tensile strength and volume resistivity initially increased with the SEUG content, which confirmed the enhancements of the chemical (HCl, NaCl, and oil) resistance properties, and then reached a maximum value at 5 wt%. When the SEUG content increased further to 10 wt%, a gradual decrease occurred, which indicated that the chemical resistance properties degraded. However, the variation of the performance after the NaOH resistance test with different SEUG contents is shown in Fig. 11. The tensile strength and volume resistivity decreased with the SEUG content, indicating that the NaOH resistance properties degraded significantly as the SEUG content was increased.

3.6 Reasonable chemical resistance promotion mechanism

Based on the results presented above, it was reasonably concluded that the HCl, NaCl, and oil resistance properties of the SEUG/ALSR composite rubbers with 5 wt% SEUG were remarkably enhanced compared to those of the neat ALSR. A possible mechanism through which the SEUG enhanced the HCl, NaCl, and oil resistances of the SEUG/ALSR composite rubbers is demonstrated in Fig. 12. The SEUG has better chemical resistance properties than ordinary ALSR due to the larger relative molecular mass, the long molecular chains, and the excellent molecular regularity. Therefore, the HCl, NaCl, and oil resistance properties of the SEUG/ALSR composite rubbers were enhanced significantly with the introduction of SEUG. Additionally, the SEUG had active Si-H groups that could react with the VPDMS during the curing process, not only resulting in the formation of closer three-dimensional cross-linked structures but also causing good dispersion in the composite rubber. The much greater crosslinking densities acted as better physical barriers to prevent the electrolytes from entering the ALSR. Based on these two factors, the chemical resistance properties initially increased with the SEUG content and reached a maximum value at 5 wt%. However, when the doped SEUG content was increased to 10 wt% in the composite rubbers, free SEUG was present (Fig. 3). The free SEUG could further form large particles due to the agglomeration, leading to an increase in the molecular chain gap and worse dispersion in the composite rubber, which

were both beneficial for the formation of diffusion channels for electrolytes. This could promote the entry of the corrosive medium into the ALSR, as illustrated in Fig. 12. Thus, these factors can explain the degradation of the chemical resistance properties for the SEUG/ALSR composite rubbers with 10 wt% SEUG.

The above analysis showed that SEUG significantly reduced the NaOH resistances of the SEUG/ALSR composite rubbers. We proposed a possible mechanism for this, as shown in Fig. 14. For the EUG, when extracted by the organic solvent extraction method, some bio-based macromolecules remained in the EUG, such as chlorogenic acid and geniposide crystals (the structures are shown in Fig. 13). As the reaction proceeded, these bio-based macromolecules were injected into the macromolecular chains of the SEUG/ALSR composite rubbers, as shown in Fig. 15. The SEUG/ALSR composite rubber chains would be destroyed by the reaction of sodium hydroxide with these bio-based macromolecules when the samples were immersed in the sodium hydroxide. Thus, the NaOH resistance of the ALSR was reduced. Furthermore, this could provide a new direction for the degradation of SEUG/ALSR composite rubbers.

Conclusions

In summary, ALSR, featuring outstanding low-dielectric properties and chemical resistance, was successfully prepared by filling it with a silicone-modified eucommia gum nano-filler. Silicone-modified eucommia gum (SEUG) was successfully prepared through a hydrosilylation reaction. As a nanofiller, the SEUG/ALSR composite rubber with SEUG contents up to 5 wt% could be easily processed. The Si-H group in the SEUG could react with VPDMS, not only resulting in good compatibility with the ALSR and preventing the SEUG from agglomeration but also leading to the formation of the much richer crosslinking densities, which was favorable for increasing the tensile strength and toughening of the ALSR. In addition, the dielectric constant of the SEUG/ALSR composite rubber were lower than 1.84, 1.79 and 1.75 at 10^2 Hz, 10^3 Hz and 10^4 Hz, respectively, which would be beneficial for decreasing the time of signal propagation in electronic devices and the long-term operation of capacitors owing to the low heat dissipation. Furthermore, because of the richer crosslinking densities and the relatively larger molecular chains, the SEUG/ALSR composite rubber containing 5 wt% SEUG showed significantly enhanced HCl, NaCl, and oil resistance performances. After 30 d of immersion in solution, the mechanical and insulation properties of the SEUG/ALSR composite rubber improved significantly compared to those of the neat ALSR. Moreover, the NaOH resistance of the ALSR was reduced by the bio-based macromolecules of the SEUG, which may provide a new direction for the degradation of SEUG/ALSR composite rubbers.

Declarations

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Tables

Table 1. Compositions of the SEUG/ALSR composite rubbers.

Component	ALSR-1	ALSR-2	ALSR-3	ALSR-4
VPDMS	100	100	100	100
PHMS-2	3.5	3.5	3.5	3.5
SEUG	0	5	10	15
1-ethynylcyclohexanol	0.1	0.1	0.1	0.1
Karsted catalyst	0.1	0.1	0.1	0.1

Figures

Figure 1

Schematic representation of the synthesis of silicone-modified eucommia gum nanofiller (SEUG).

Figure 2

FT-IR spectra of SEUG and EUG.

Figure 3

Cross-sectional SEM images of (ALSR-1) neat ALSR, (ALSR-2) SEUG/ALSR composite rubber with 5 wt% SEUG content, (ALSR-3) SEUG/ALSR composite rubber with 10 wt% SEUG content, and (ALSR-4) SEUG/ALSR composite rubber with 15 wt% SEUG content.

Figure 4

Tensile strength and elongation at break of SEUG/ALSR composite rubbers with different contents of SEUG.

Figure 5

Shore A hardness values of the SEUG/ALSR composite rubbers with different contents of SEUG.

Figure 6

Dielectric constants of the SEUG/ALSR composite rubbers with different contents of SEUG.

Figure 7

Possible mechanism through which the SEUG affected the low-dielectric properties of the ALSR.

Figure 8

Tensile strength and volume resistivity of the SEUG/ALSR composite rubbers after HCl resistance tests.

Figure 9

Tensile strength and volume resistivity of the SEUG/ALSR composite rubbers after NaCl resistance tests.

Figure 10

Tensile strength and volume resistivity of the SEUG/ALSR composite rubbers after oil resistance tests.

Figure 11

Tensile strength and volume resistivity of the SEUG/ALSR composite rubbers after NaOH resistance tests.

Figure 12

Possible mechanism through which the SEUG enhanced the HCl, NaCl, and oil resistances of the ALSR.

Figure 13

Structures of bio-based macromolecules remaining in the EUG.

Figure 14

Possible mechanism through which the SEUG reduced the NaOH resistance of the ALSR.

Supplementary Files

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- [Scheme1.jpg](#)