Use of gradient laminating to prepare NR/ENR composites with excellent damping performance
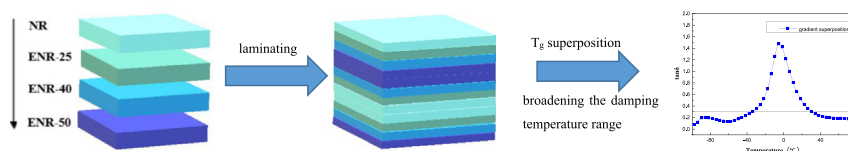
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HIGHLIGHTS

- A novel processing method has been developed to broaden the damping temperature range of natural rubber.
- Good compatibility between the layers and superposition of gradient T_g broaden the damping temperature range.
- From −24.8 °C to 44.9 °C, the results exhibit a broad range of effective damping ($\tan\delta_{min} > 0.3$) temperature.

GRAPHICAL ABSTRACT

ABSTRACT

A novel processing method has been developed to broaden the damping temperature range of natural rubber (NR). Natural rubber and three kinds of epoxidized natural rubber (ENR, epoxidized degree: 25%, 40%, 50%) are arranged in a certain order to prepare NR/ENR-25/ENR-40/ENR-50 quaternary layered gradient material in a laminating way. By utilizing the relatively narrow T_g difference and the high compatibility between NR and ENR series, results show that the presence of three kinds of ENR can effectively broaden the effective damping temperature range ($\tan\delta > 0.3$) of NR. Specifically, the prepared NR/ENR-25/ENR-40/ENR-50 layered gradient material has the best damping performance due to the T_g superposition when the order of gradient superposition of epoxidized degree is adopted. Furthermore, the peak value of the loss factor ($\tan\delta_{max}$) of layered gradient materials increases with the increment of layers, and when the layer number stands at 32, the peak of $\tan\delta$ value can be as high as 1.815, covering an effective damping temperature range of 69.7 °C from −24.8 to 44.9 °C ($\tan\delta > 0.3$).

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1. Introduction

In recent years, the rapid development of science and technology and the iterative updating of industrial equipment have put forward higher demands on engineering materials. The damping materials used for vibration and noise reduction are no exception [1–6]. Among the existing damping materials, polymeric viscoelastic materials have the high damping loss factor and have been widely used in a broad base of fields [7–10]. At present, many researches on polymer damping material focus on the butyl rubber and its halides. Such rubber has a large density of methyl side and its unique liquid transition as well as glass transition imparts it good damping properties, but poor processing performance at the same time [11–13]. In contrast, natural rubber (NR), the earliest used rubber, is rarely used for damping materials due to its low T_g and poor damping performance at room temperature and above. However, compared with other rubber species, NR has excellent mechanical strength, processing performance and low temperature damping properties. Naturally, the modification of NR and its widely applications in damping field are of great research significance and have real economic value [14,15].

In general, the damping temperature range of a single rubber species is generally narrow, about 20–30 °C around T_g and mostly below room temperature. But in practical applications, the damping material needs

to have a loss factor (tanδ) that >0.3 over a temperature range of >60 °C–80 °C which include the room temperature and above the temperature. So it is often necessary to modify the rubber to improve its damping properties [16]. Epoxidized natural rubber (ENR), modified by NR latex, retains the basic structure of NR and has good mechanical properties. In the presence of epoxy groups, the polarity of the ENR increases when compared to NR and the T_g of ENR increases with higher epoxidized degree. So we suggest that the alike but different structure of ENR and NR would make a big difference when combined with each other [17,18].

Typically, there are several methods to improve the damping property of polymer, such as blending [19–21], copolymerization [22–24], interpenetrating network structure [25,26], hybridization of organic small molecules [27], the introduction of the dangling chain [28,29] and so on. Herein, unlike the traditional modification methods, we propose a new idea of using the lamination method to obtain quaternary layered damping material with high damping properties. Three types of ENR rubber (ENR-25, epoxidized degree of 25%, ERN-40: epoxadatized degree of 40%, ENR-50: epoxidized degree of 50%) were used in combination with NR, and the epoxidation gradient superposition theory was also applied by utilizing the different epoxidized degree. A NR/ENR-25/ENR-40/ENR-50 quaternary layered graded damping material was prepared by laminating to achieve the epoxidized degree sequence of the adhesive layer from small to large and then from large to small, so as to make the most of the T_g difference as well as the good compatibility among each component together with the inter-facial effect between various layers, bringing about high damping materials with much broadened damping temperature range and practical damping value.

Experiments show that, within NR/ENR-25/ENR-40/ENR-50 quaternary layered damping materials, the molecular structures of molecular chains are similar in adjacent layers with epoxidized gradient superimposition. Accordingly, the polarity is relatively small and the compatibility is so good that the free volume between molecular chains can be shared. As a result, the layered structure can effectively improve the damping performance near room temperature. What's more, the material overcomes the poor mechanical property of traditional layered materials. When adding 40 phr filler and 12 layers, the tensile strength of the material can reach 12.1 MPa and the elongation at break is 447% with an effective damping temperature up to 76.6 °C, ranging from −32.3 °C to 44.3 °C, which is sufficient enough to meet the performance requirements of damping NR in engineering applications.

2. Experiment details

2.1. Materials

The NR used in this research was a standard rubber (3 L), and was supplied by Guang Zhou Liben Rubber Raw Materials Co., LTD, China; Epoxidized Natural Rubbers (ENR, extents of epoxidation: 25%, 40%, 50%), were manufactured by China Tropical Agricultural Science Institute of Agricultural Products Processing, China; Carbon black (high abrasion furnace, HAF-N330) was manufactured by Cabot Corporation, USA; Precipitated silica (Rhodia 142) was manufactured by Luodiya Silica White (qingdao) Co., Ltd.; Rubber additives such as zinc oxide (ZnO), stearic acid, basic magnesium carbonate (MgCO3), N cyclohexyl 2 benzothiazole sulfenamide (CZ), 2,2′ dibenzothiazoledisulfde (DM), Butylated phenol formaldehyde resin (2402PF), sulfur (S8) used in the study were supplied by Guangzhou Jin Changsheng Technology Corporation, China, and were of commercial grade.

2.2. Preparation of multi-components blends and multi-components layered damping material

1) NR, ENRs gross rubbers were produced according to the formulas showed in Table 1, respectively. The raw rubbers were first

Table 1
The details of basic formula.

| Ingredient (phr)	Formulas			
	NR	ENR-25	ENR-40	RNE-50
Raw rubber	100	100	100	100
MgCO3	–	2	2	2
ZnO	5	5	5	5
Stearic acid	2	2	2	2
DM	0.5	0.5	0.5	0.5
CZ	1.5	1.5	1.5	1.5
S8	1.5	–	–	–
2402PF	–	10	10	10

masticated on a two-roll mill for 3 min at ambient temperature, then rubber additives were added and mixed for another 7 min. Compounded rubbers of NR, ENRs were obtained after that.

2) The NR/ENR-25/ENR-40/ENR-50 multi-component blends were kneaded based on NR/ENR-25/ENR-40/ENR-50 raw rubber mass ratios of 40/25/15/20, 50/10/10/30, 50/15/15/20, 55/5/15/25, 55/15/15/15, 60/20/10/10 respectively. Those blending samples were turned into sheets and then were pressed under the pressure of 100 MPa for 25 min at 160 °C.

3) multi-components layered damping material: The compounded rubbers of NR, ENR-25, ENR-40, ENR-50 were rolled out with the same roll space by open mill. After rolling out, they were cut into rubber sheets with the same shape and weight. These rubber sheets were stacked in a certain order to form multi-layer mixed rubbers. First, the mixed rubbers were put into a 2–4 mm mould and were pressed at room temperature, and then initial cured in the vacuum oven under the pressure of 0.1 MPa for 8 min at 160 °C, and finally second cured on the vulcanizing press under the pressure of 100 MPa for 17 min at 160 °C.

2.3. Major performance testing and characterization

2.3.1. Dynamic mechanical analysis (DMA)

The specimens with dimensions of $10 \times 6 \times 2$ mm were subjected to dynamic mechanical analysis (DMA 242C, NETZSCH) by using a mode of tensile mode. A frequency of 10 Hz and a dynamic force on sample of 4 N were used for testing samples. The specimens were scanned from −110 °C to 80 °C at a heating rate of 4 °C/min.
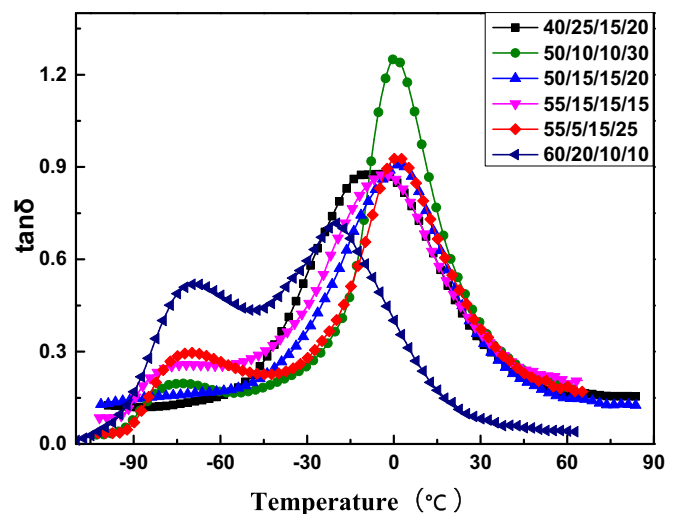
Fig. 1. The tanδ-T curves of NR/ENR-25/ENR-40/ENR-50 blends with different blend ratios (10 Hz).