Research Article

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Preparation and properties of natural rubber composite with CoFe_2O_4 -immobilized biomass carbon

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Abstract: Designing versatile rubber as a multifunctional elastomer is of great importance, incorporating it with biomass-derived nanoblocks will mitigate environmental challenges. Here biosynthesized natural rubber (NR) composites with CoFe_2O_4 -immobilized biomass carbon (BC) derived from macadamia nutshells were fabricated by facile mechanical mixing. Morphological analysis indicates that CoFe_2O_4 nanoparticles are uniformly anchored on the surface of BC, forming intact electromagnetic loss networks in NR matrix. As a consequence, the as-fabricated NR/ CoFe_2O_4 @BC composites demonstrate enhanced mechanical, thermal, and electromagnetic performance. Particularly, NR/ CoFe_2O_4 @BC composite shows the best microwave attenuation capacity when CoFe_2O_4 @BC loading is 40 phr, with the minimum reflection loss (RL) of -35.00 dB and effective absorption bandwidth (RL < -10 dB) of 1.60 GHz. All results indicate that this work open new paradigm for multiple applications based on biosynthetic elastomer with the sustainable biomass derived nanoblocks.

Keywords: natural rubber, biomass carbon, mechanical mixing, microwave absorbing performance, composite

1 Introduction

Due to their light weight, low cost, flexibility, and other advantages, elastomer has attracted a tremendous surge of interest among researchers (1). In order to acquire robust enhancement in mechanical, optical, electrical, magnetic, and thermal properties, elastomer is always modified with nanofillers (2). Over the past decades, extensive efforts have been made for improving the property of rubber by incorporating numerous fillers, including carbon black (3), carbon nanotube (4), and graphene (5). However, most of the current elastomeric composites are made of fossil-derived matrix and nanofillers, where the emission of pollutant gasses is inevitable. With the increasing environmental awareness and growing demand for sustainability, new generation elastomeric composites should be ideally based on sustainable raw materials using low-cost processing and without any health issues.

To minimize the environmental impacts caused by fossil-derived elastomeric composites, an alternative is incorporating natural rubber (NR) with biomass-derived nanofillers (6). Extracted in the form of latex from the Hevea Brasiliense tree, NR demonstrates excellent physical and chemical properties (7), such as elasticity, flexibility, durability, etc. Aim to reinforced mechanical property, several studies have been reported for the preparation of NR nanocomposites with biomass-derived nanofillers. However, limited by complicated and incompatible fabrication compounding methods, few materials can be designed and developed to simultaneously meet structural and functional requirements in some specific applications.

Here a kind of sustainable material based on NR composite with CoFe_2O_4 -immobilized biomass carbon (BC) derived from macadamia nutshells was fabricated by facile environment-friendly approach. The obtained composites

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were characterized by different techniques, and the morphology, rheology, mechanical, thermal, and electromagnetic properties of as-fabricated NR/ CoFe_2O_4 @BC composites were systematically investigated. In addition, a possible model is proposed to reveal morphology associated microwave attenuation mechanism.

2 Materials and methods

2.1 Materials

Macadamia nutshells were provided by South Subtropical Crop Research Institute, China Academy of Tropical Agricultural Sciences. Natural rubber latex (NRL) with a solid content of 60% was sourced from Shuguang State Rubber Farm (Zhanjiang Xiashan Xinjia Rubber & Plastic Products Co., Ltd). High purity argon (>99.999%) was supplied by Zhanjiang Oxygen plant. $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$, $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$, and ammonia solution were supplied by Shanghai Aladdin Technology Co., Ltd. Rubber compounding chemicals including zinc oxide (ZnO), stearic acid (SA), zinc diethyl dithiocarbamate (ZDC), and sulfur (S) were of analytical grade and used as received.

2.2 Preparation of CoFe_2O_4 @BC

CoFe_2O_4 @BC was prepared by hydrothermal and carbonization. Briefly, BC was prepared by pyrolyzing macadamia nutshells in tubular furnace at 400°C for 2 h with a ramp rate of 5°C·min⁻¹ in argon atmosphere. Then, 1.8 g BC were added into 50 mL of aqueous solution with 4 g $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ and 1.5 g $\text{Co}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$, accompanying with ultrasonication for 10 min. Subsequently, ammonia solution was slowly dripped until pH reached 9. Thereafter, the mixture was transferred into a Teflon-lined autoclave of 100 mL, and the hydrothermal reaction was carried out at 200°C for 8 h. After cooling to room temperature, black CoFe_2O_4 @BC powder was obtained by washing with deionized water and vacuum drying (60°C, 12 h).

2.3 Fabrication of NR/ CoFe_2O_4 @BC composites

NR/ CoFe_2O_4 @BC composites were prepared according to the following procedures. 150 g NRL was mixed with 4 mL of compounding agent suspension containing 1.1 g S, 2.7 g ZnO, and 2.2 g ZDC. Then, 30 mL of sonication

treated CoFe_2O_4 @BC suspension with different concentrations were added into the above latex mixture of 20 g and stirred for 30 min. Subsequently, the as-prepared mixture was frozen by -196°C liquid nitrogen and dried at -92°C for 72 h. Subsequently, the master-batch was masticated by the two roll-mill at 70°C for 3 times. Finally, the dried rubber masterbatches were placed in a mold (140 mm × 136 mm × 1 mm) and vulcanized at 145°C for the optimum cure time (t_{90}) that was deduced from the curing curves (MDR 2000E). The obtained composites were designated as NR/ CoFe_2O_4 @BC- x , in which x represents the CoFe_2O_4 @BC content. For example, NR/ CoFe_2O_4 @BC-10 means a NR composite with 10 phr CoFe_2O_4 @BC. In comparison, the NR was prepared by the same process in the absence of CoFe_2O_4 @BC.

2.4 Characterization

X-ray diffraction (XRD) patterns were recorded on a Bruker D8 Advance (Bruker Co. Ltd, Germany), with scanning speed of 2°·min⁻¹. Raman spectra were recorded by the confocal Raman spectrometer (HORIBA JY Lab RAM HR Evolution, France). Morphology was visualized by Scanning Electron Microscopy (SEM) (Hitachi, S4800, Japan). Energy-Dispersion X-ray spectroscopy (EDAX, USA) was used for elemental analysis. The storage shear moduli (G') of the rubber compounds with curatives was evaluated by using a Rubber processing analyzer (RPA 2000, Alpha Technologies, USA) at a temperature of 100°C, frequency of 0.5 Hz, and varying strains in the range of 0.28–100%. The vulcanization characteristics were analyzed with a Rotorless Curemeters (MDR2000E, Wuxi, P.R. China). The thermal stabilities of the tested composites were investigated using a simultaneous thermal analyzer (STA449/4, NETZSCH Instruments, Germany) under N₂ atmosphere. Dynamic mechanical analysis was performed with a dynamic mechanical analyzer (DMA, TA-Q800, USA). Volume electrical conductivity (σ) was measured by using a ZC-68 high resistance meter (Shanghai, P.R. China). The electromagnetic parameters were obtained by using a vector network analyzer (Agilent, N5244A, USA) in the frequency range of 2–18 GHz.

3 Results and discussion

3.1 Nanostructure of CoFe_2O_4 @BC

Figure 1a shows the XRD patterns of BC and CoFe_2O_4 @BC. It can be seen that there are two broad diffraction peaks

Figure 1: (a) XRD patterns and (b) Raman shifts of BC and CoFe₂O₄@BC.

located at 24.4° and 43.9° for BC, which are assigned to the (002) and (100) crystal planes of graphitic carbon, respectively. For CoFe₂O₄@BC, while the additional characteristic peaks at 23.16° and 34.00° are corresponding to the (006) and (012) planes of the spherical structure of CoFe₂O₄, the major characteristic peaks located at $2\theta = 35.4^\circ$, 43.1° , and 62.6° are assigned to the (311), (400), and (440) planes of spinel-type CoFe₂O₄ (JCPDS No. 22-1086) (8). From the Raman shifts shown in Figure 1b, CoFe₂O₄@BC demonstrates two main peaks at $\sim 1,373$ and $\sim 1,585$ cm⁻¹, which are derived from the D-band and G-band of BC, respectively. It is well-known that

the D-band stands for amorphous or disorder carbon and G band means graphitization carbon, and the intensity ratio of the D band and G band (I_D/I_G) can be applied to assess the disorder and defects of the carbon atoms. The I_D/I_G of CoFe₂O₄@BC is about 1.07, which is higher than that of BC (0.86), indicating that CoFe₂O₄@BC has more lattice defects (9).

The morphology of the as-synthesized CoFe₂O₄@BC was visualized by using SEM-EDS and Transmission electron microscopy (TEM). As shown in Figure 2a, CoFe₂O₄ nanocrystals with size of 0.1–5 μm were uniformly anchored on the surface of BC-based three-dimensional (3D) interconnected

Figure 2: (a) SEM image and (b–e) elemental mapping of C, Fe, Co, O of CoFe₂O₄@BC. (f) TEM image of CoFe₂O₄@BC.

architecture (10). In addition, the elemental mappings in Figures 2b–e display the distribution of C, Fe, Co, and O elements on the surface of the as-synthesized sample, further confirming the successful synthesis of CoFe_2O_4 @BC. TEM measurements were performed to investigate the inner structure of CoFe_2O_4 @BC. As observed in Figure 2f, numerous nanopores can be observed in amorphous BC, and the as-generated CoFe_2O_4 nanoparticles are randomly distributed in the porous carbon, which is in good agreement with the conclusion obtained from the analyses on SEM results.

3.2 Morphology of NR/ CoFe_2O_4 @BC composites

Morphology characterization is also performed to investigate the distribution of CoFe_2O_4 @BC. As shown in Figure 3a, the dense packed NRL fully coalesce into a smooth surface, where the white dots may be attributed to the ZnO particles added during the curing process. While NR/ CoFe_2O_4 @BC-10 and NR/ CoFe_2O_4 @BC-20 exhibit good filler dispersion, large aggregations are observed in NR composites when the CoFe_2O_4 @BC loading is more than 30 phr. As indicated in Figure 3b and c, some unconnected CoFe_2O_4 @BC and coalesced NR region are formed in NR composites with low filler loading. When the amount of CoFe_2O_4 @BC increases to 30 phr, the filler network becomes denser and cause a connect pathway throughout the rubber matrix. However, for NR/ CoFe_2O_4 @BC-40 and NR/ CoFe_2O_4 @BC-50, a large number of CoFe_2O_4 @BC particles appear to be pulled

out from the rubber matrix (Figure 3e and f). At this stage of higher concentration, the aggregation of CoFe_2O_4 @BC causes the filler–elastomer interaction to decrease, thus deteriorating the characteristic properties of the resulted composites.

3.3 Rheological behavior of NR/ CoFe_2O_4 @BC composites

It is well accepted that the formation of a filler network takes place when fillers are incorporated in elastomer matrix due to their tendency to agglomerate resulting from filler–filler interactions, which shows important effect on the final properties of the composites. Figure 4a displays the influence of the dynamic strain amplitude on the storage modulus (G') of NR/ CoFe_2O_4 @BC composites at different filler loadings. It can be seen that the G' is at the highest at small amplitude and gradually decreases to a lower value, which is described as “Payne effect” (11). Moreover, the magnitude of the Payne effect increases with the increase in the CoFe_2O_4 @BC content, indicating a higher filler network formation and worse particle dispersion (12). To investigate the influence of filler loading on the vulcanization kinetics of NR/ CoFe_2O_4 @BC, the curing curves of resulted composites are shown in Figure 4b. Obviously, the curing curves are systematically shifted toward the short time side with the increase in CoFe_2O_4 @BC loading, implying that the vulcanization process of NR is markedly accelerated. Moreover, the presence of CoFe_2O_4 @BC elevates both

Figure 3: SEM images of NR composites with different CoFe_2O_4 @BC loadings: (a) 0 phr, (b) 10 phr, (c) 20 phr, (d) 30 phr, (e) 40 phr, and (f) 50 phr.

Figure 4: (a) Nonlinear dynamic and (b) curing curves of NR composite with different CoFe₂O₄@BC loadings.

the minimum and maximum torque values of the curing curves. As the filler content increases, the curing rate of NR/CoFe₂O₄@BC is slower. The curing rate of NR gum is the fastest ($t_{90} = 13.7$ min), and that of the other composite ranges from 16 to 20 min, implying that the addition of CoFe₂O₄@BC greatly prolongs the crosslinking reactions.

3.4 Mechanical properties of NR/CoFe₂O₄@BC composites

It is well-known that the mechanical performance of a composite generally depends on the various filler parameters including geometry, stiffness, and dispersion (13). Table 1 summarizes the mechanical properties of NR/CoFe₂O₄@BC composites. Evidenced by the increased modulus at 100%, 300%, and 500%, the incorporation of CoFe₂O₄@BC improves the mechanical properties of NR. However, the tensile strength and elongation at break of NR/CoFe₂O₄@BC decrease gradually, which is ascribed to

the rigid CoFe₂O₄@BC networks serving as electromagnetic loss pathway and nonelastic reinforcement that can cause the brittleness and ultimate failure of the composites as stress concentrative points. This phenomenon agrees well with the fracture micromechanism of inorganic particles and the electrical conductivity of conductive filler-filled polymers.

In order to further understand the reinforcement mechanism of CoFe₂O₄@BC, dynamic mechanical measurements were performed at 10 Hz as a function of temperature (100–100°C). It can be seen from Figure 5a that the storage modulus (G') suddenly drops down by three orders of magnitude with the increase in temperature, which is corresponding to the glass-rubber transition that ascribed to chain motion-induced energy dissipation (14). Comparing with that of neat NR, the storage modulus of NR/CoFe₂O₄@BC increased by a factor of 1.25–1.75. Figure 5b shows the response relationship between the loss factor ($\tan \delta$) and the temperature of different CoFe₂O₄@BC fillings, of which all composites have peaks in the range from -38°C to -45°C , which represent the glass transition peak

Table 1: Mechanical property of NR and its composites with CoFe₂O₄@BC

Samples	Tensile strength (MPa)	Modulus (MPa)			Elongation at break (%)
		100%	300%	500%	
NR	16	1	1	3	763
NR/CoFe ₂ O ₄ @BC-10	11	1	2	3	719
NR/CoFe ₂ O ₄ @BC-20	7	1	2	4	598
NR/CoFe ₂ O ₄ @BC-30	8	1	2	7	526
NR/CoFe ₂ O ₄ @BC-40	5	1	2	—	461
NR/CoFe ₂ O ₄ @BC-50	4	2	3	—	410

Figure 5: Temperature depended (a) storage modulus and (b) $\tan \delta$ of NR composite with different CoFe₂O₄@BC loadings.

of NR composites. Meanwhile, the maximum value of $\tan \delta$ decreases with the increase in the CoFe₂O₄@BC content, which is usually observed in reinforced elastomeric composites and interpreted as a decrease in chain mobility due to interaction with nano-fillers.

3.5 Thermal properties of NR/CoFe₂O₄@BC composites

Thermal properties of NR composites with different phr of CoFe₂O₄@BC were investigated by differential scanning calorimetry (DSC) and thermogravimetric analysis (TGA) and are shown in Figure 6. It is observed from Figure 6a that the glass transition temperature (T_g) of pure NR gum

compound is -61.3°C . In addition, due to the strong interfacial interaction, T_g of NR/CoFe₂O₄@BC composites slightly increases with the increase in the CoFe₂O₄@BC, ranging from -61°C for NR/CoFe₂O₄@BC-10 to -59°C for NR/CoFe₂O₄@BC-50. The TGA curves of NR/CoFe₂O₄@BC composites are shown in Figure 6b. It can be seen that all composites demonstrate a dominant weight loss process around 380°C , which is ascribed to the decomposition of the rubber chains. The amounts of residue for all the NR/CoFe₂O₄@BC composites are higher than that of NR, which is attributed to the CoFe₂O₄@BC-based fillers that remain after the thermal treatment. As expected, the residue increases with the increase in the filler content. Particularly, the total mass loss for NR10–NR50 composite materials is 91.7%, 83.4%, 77.6%, 73.0%, and 66.5%, respectively.

Figure 6: (a) DSC and (b) TG curves of NR composite with different CoFe₂O₄@BC loadings.

Figure 7: Electrical conductivity of NR composites with different $\text{CoFe}_2\text{O}_4@\text{BC}$ loadings.

3.6 Electromagnetic properties of NR/ $\text{CoFe}_2\text{O}_4@\text{BC}$ composites

Figure 7 shows that the electrical conductivity of NR/ $\text{CoFe}_2\text{O}_4@\text{BC}$ composites is a function of $\text{CoFe}_2\text{O}_4@\text{BC}$ loading. It can be seen that the electrical conductivity of NR is about $8.0 \times 10^{-13} \text{ S}\cdot\text{m}^{-1}$, implying good insulating property. With the increase in $\text{CoFe}_2\text{O}_4@\text{BC}$ content, a sudden increase in the conductivity ranging from $1.2 \times 10^{-10} \text{ S}\cdot\text{m}^{-1}$ for 10 phr to $5.6 \times 10^{-9} \text{ S}\cdot\text{m}^{-1}$ for 50 phr

was observed, indicating the formation of a preliminary conductive network (15).

The microwave absorbing performances of the materials are first evaluated by using the reflection loss (RL) values as an indicator. Based on the transmission line theory, RL values can be calculated using the following equations (16,17):

$$\text{RL} = 20 \lg \left| \frac{Z_{\text{in}} - 1}{Z_{\text{in}} + 1} \right| \quad (1)$$

$$Z_{\text{in}} = \sqrt{\frac{\mu_r}{\epsilon_r}} \tanh \left(j \frac{2\pi f d}{c} \sqrt{\mu_r \epsilon_r} \right) \quad (2)$$

where Z_{in} is the input impedance of the absorber, μ_r is the relative complex permeability, f is the frequency of the electromagnetic wave, c is the light velocity in the free space, and d is the absorber's thickness. Figure 8 depicts contour plots of frequency and thickness depended RL NR composites with different $\text{CoFe}_2\text{O}_4@\text{BC}$ loadings. It can be observed that the matching frequency of RL_{min} shifts gradually towards lower frequency with increase in the absorber thickness, which can be explained according to quarter wavelength matching model. The minimum of RL_{min} of NR is -2.76 dB , implying relatively poor microwave absorption performance. The microwave absorbing performance of NR composites is largely enhanced by introducing more $\text{CoFe}_2\text{O}_4@\text{BC}$. For instance, NR/ $\text{CoFe}_2\text{O}_4@\text{BC}$ -30 shows a RL_{min} of -29.00 dB at 17.50 GHz , which is 10.5 times that of NR composite. When the filler loading is

Figure 8: Contour plots of frequency- and thickness-depended RL NR composites with different $\text{CoFe}_2\text{O}_4@\text{BC}$ loadings.

Figure 9: Schematic illustration of the microwave attenuation mechanism of NR/CoFe₂O₄@BC composite.

40 phr, NR/CoFe₂O₄@BC composite exhibits strongest microwave attenuation capacity, with $RL_{\min}$ of -35.00 dB and effective absorption bandwidth ($RL < -10$ dB) of 1.60 GHz. Moreover, this NR/CoFe₂O₄@BC can be explored as a practical absorber in a frequency range of 14.98–18.00 GHz via tuning of the thickness.

Sustainable development requires products to be prepared by simple methods with renewable and environmentally friendly sources. Figure 9 schematically illustrates the probable microwave attenuation mechanism of NR/CoFe₂O₄@BC. Initially, BC possessed numerous defects and porous structures, which can bring about conduction loss, dipole, and interfacial polarization. Then, the introduction of CoFe₂O₄ consumes a large proportion of the electrical energy due to its resistance, resulting in excellent magnetic loss. Moreover, the interfaces between CoFe₂O₄, BC, and NR can also work as the polarization centers, which could generate excessive polarization relaxations. Additionally, the highly porous structures of BC as well as CoFe₂O₄@BC-based electromagnetic loss network in NR matrix can also bring about the multiple reflections/scaterings, which can further enhance the microwave absorption performance (18). Therefore, the best impedance matching together with large attenuation capability endows NR/CoFe₂O₄@BC composites with high absorbing performance.

4 Conclusion

In summary, NR/CoFe₂O₄@BC composites with enhanced mechanical, thermal, and electromagnetic performances were fabricated by a facile, efficient, and environmentally friendly approach. Particularly, the incorporation of

CoFe₂O₄@BC ensures a higher electromagnetic loss network in NR matrix, endowing the resulted composites with balanced impedance matching and strong attenuation capability. Therefore, NR/CoFe₂O₄@BC with a filler loading of 40 phr demonstrates enhanced microwave absorbing performance, with the minimum RL of -35.00 dB and effective absorption bandwidth ($RL < -10$ dB) of 1.60 GHz. Moreover, the addition of CoFe₂O₄@BC increases the mechanical, thermal, and electrical properties of the NR composites. Taking the fabrication approaches, raw materials, and ultimate properties into consideration, the as-fabricated NR/CoFe₂O₄@BC composite will be a promising candidate as sustainable multifunctional elastomeric materials.

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