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The Effect of Intermolecular Interactions on the Properties of Poly(chlorobutyl) / Graphene Oxide and Nanoclay Nanocomposites

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ABSTRACT

Nano composites of graphene oxide and modified nanoclay (Cloisite 15A) were produced based on 80 phr chlorobutyl and 20 phr natural rubber according to inner liner recipe. The results show modified nano fillers have remarkable change in dispersion in the matrix. Subsequently Improved cured products properties. Exfoliation of graphen oxide and Cloisite 15A was confirmed by XRD and TEM. In the case of graphite intercalation has happened. Also physical and mechanical properties of this composite was studied.

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1. Introduction

Since 1984, when the Japanese research team of Toyota developed polyamide 6/nanoclay, nanocomposites became one of the important issues [1]. The high aspect ratios, high surface area, and enhancement of significant nanoparticle properties compared to conventional fillers have attracted the attention of many researchers to the development, identification and modeling of polymer nanocomposites [2-3]. The key issue in nanocomposite production is to achieve the highest inter-surface compatibility and complete dispersion of filler in the matrix. Single-layered layers of two-dimensional nanosheets have a higher aspect ratio than their microscopic aggregates. Therefore, nanoscale lamination is required to achieve the highest nanocomposite efficiency [4-10]. Due to the high contact surface and the amount of interactions with the polymer matrix in unit volume, the nanoclay has a significant increase in physical-mechanical properties and decreases in gas permeability in elastomeric nanocomposites [4-5]. The amount of dispersion of modified nanoclay in the rubber matrix depends essentially on the nature of the clay cluster structure [5], the mixing condition [6], (shear rates of mixing and temperature), and the polarity of the rubber matrix [6]. The elastomers exhibit high viscosity during the mixing process due to their high molecular weight, which causes the shear stress required to delaminate the silicate layers of nanocaly [4,6].

In addition to clay nanocomposites, many researchers are working on improving the mechanical, electrical, and barrierity properties of rubber / graphene nanocomposites [6-10]. Graphene is a two-dimensional layer consisting of carbon atoms with sp2 hybrid that are arranged in the structure of the honeycomb that placed on each other [7]. Graphene has an electrical conductivity, mechanical flexibility, optical clarity, excellent thermal conductivity and a small coefficient of thermal expansion, which has made it an excellent attraction among researchers and industrials. Due to its exceptional properties, graphene has been used as an ideal material for the electronics industry, such as battery connections, superstructures, solar cells, sensors, composites, gas separation membranes, hydrogen storage, and biological sensors. Graphene has similar properties in comparison to nanotubes, but its larger surface area can be considered as open-ended nanotubes. In addition, the cost of producing graphene sheets is very low compared to nanotube costs and can be as a cheap alternative to apply in nanocomposites [8-9]. Sheets of graphene oxide can easily be dissipated in water because it has hydroxyl groups. The use of graphene oxide in reinforced composites is one of its applications, it can greatly improve the mechanical and thermal properties of polymer matrix. Zachariah et al. [4] reported improvements in the properties of natural rubber and Chlorobutyl composites by increasing the different nanoparticles of modified clay and provided a mechanism for the of dispersion of these nanoparticles. Tiwari et al. investigated the rheological properties of increasing the graphite nanoparticles to chlorobutyl matrix in the range of -100 °C to 100°C to investigate the effect of the glass transition temperature on the particle dispersion [6]. Another report from the same group concentrates on the effect of nano graphite particles enhancement on the physical mechanical properties of chlorobutyl matrix [7]. In other papers, the same group studied the effect of multi wall carbon nanotubes on chlorobutyl rubber and the distribution of these particles in high percentages by scanning electron microscopy [8-9].

The properties of the chlorobutyl rubber and the butyl rubber properties are very similar to each other. An increase of about 2.1% by weight of chlorine to butyl rubber to increase the reaction of butyl groups in polymer without changing the number of them to enhance the possibilities of vulcanization, and thus the presence of double bond and chlorine in these rubber present different way of vulcanization. The most important consumption of chlorobutyl rubber is in the manufacture of inner liner of tire and is also used in the manufacture of rubber and rubber products, as well as non-toxic vulcanization, it is also used in the manufacture of medical and food applications. It also used in glue coatings, bunker parts and heat resistant parts up to 150 °C Chlorobutyl rubber reinforced with nano fillers has been shown to be remarkably improved. in physical-mechanical properties [4,7] and reduction of gas permeability have been reported in the references [10]. These properties increase due to the entry of nano-fillers with an impermeable sheet structure. So produce nanocomposites that cover a wide range of products from packaging, pharmaceuticals and automotive industries.

Many researchers have done extensive research on the use of conventional nano-fillers, but the comparison of the properties of these additives has not been studied at the same time. In this report, a comparison was made between graphite nanoparticles, graphene oxide and nanoclay (Cloisite 15A) as nanofillers and chlorobutyl rubber as matrix using X-ray diffraction techniques, transition electron microscopy (TEM) and tensile analysis.

2. Experimental Section

2.1. Materials

Chlorobutyl rubber produced by Langus Company of Belgium and Natural Rubber (SMR20) from Malaysia, the Cloisite 15A clay from Southern, graphite, sulfuric acid, sodium nitrate, potassium permanganate, polymeric sulfur (S8), n oxyethylene 2 -benzothiazole Sulfenamide, zinc oxide and stearic acid were purchased from Merck.

2.2. Production of Graphene Oxide

Based on the Hummers & Offeman method [11], 3 g of graphite with 3 g of sodium nitrate and 200 ml of sulfuric acid (98%) were mixed in 1000 ml balloons in an ice bath equipped with a circulator. Then 9 g of potassium permanganate was added to the above mixture. The solution was kept at 35 °C for 18 hours. The mixer was used for uniformity throughout the reaction. At the end of the reaction, the solution was washed with oxygenated water and distilled water then mixture centrifuged and dried.

2.3. Nanocomposites production

In order to improve the mixing of compound 20phr of natural rubber (SMR20) was used. A two-liter banbury mixer was used to mix the compounds. Compositions of the compounds were shown in Table 1. The rheometer (ODR2000-alpha) was used to obtain optimal curing conditions (scorch time) for each of the compounds. Specimens for tensile test were prepared by compression molding of produced compounds and cutting them in dumbbell shaped specimens with 2mm thickness. (Figurer1) shows four samples prepared for the tensile test.

Table 1. Compositions of the compounds.

LNG HE Name	CIIR/NR-C 15A	CIIR/NR-graphen oxide	CIIR/NR-graphite	CIIR/NR
Ingredients (phr)				
CIIR	80	80	80	**80**
NR	20	20	20	**20**
Graphite	-	-	5	**-**
Graphene-oxide	-	1	-	**-**
Cloisite 15A	4	-	-	**-**
ZnO	3	3	3	**3**
stearic acid	2	2	2	**2**
S8	3	3	3	**3**
N-oxydiethylene-2-benzothiazole sulfonamide	2	2	2	**2**

Figure 1. Cured samples from left, CIIR/NR, CIIR/NR-graphite, CIIR/NR-graphen oxide, CIIR/NR-C 15A

2.4. Characterization

Dumbbell-shaped specimens were prepared from the cured compounds then tensile analysis were done by INSTRON machine at a speed of 10 mm / min. The X-ray diffusion test was carried out using an X-ray diffraction device manufactured by PHILIPS with a copper anode and λ=1/541 at ambient temperature. To analyze quality of nanoparticles distribution in the matrix, the TEM observations were carried out using a Zeiss -EM 900 (80 keV) electron microscopy.

3. Results and Discussion

The XRD spectra obtained from graphene nanocomposites and clay 15A clay exhibit a good distribution of nanoplates (Figurer 2), while in the case of CIRR / NR-graphite composite, delaminating state did not occur, However, the peak has been moved to lower 2θs and has not been eliminated due to the intercalation of graphite layers. Therefore, it is possible to clearly observe the effect of the compatibility of clay and graphene nanoparticles. In order to verify delaminating of CIIR / NR-graphene oxide sample, electron microscopic images were used (Figurer 3).

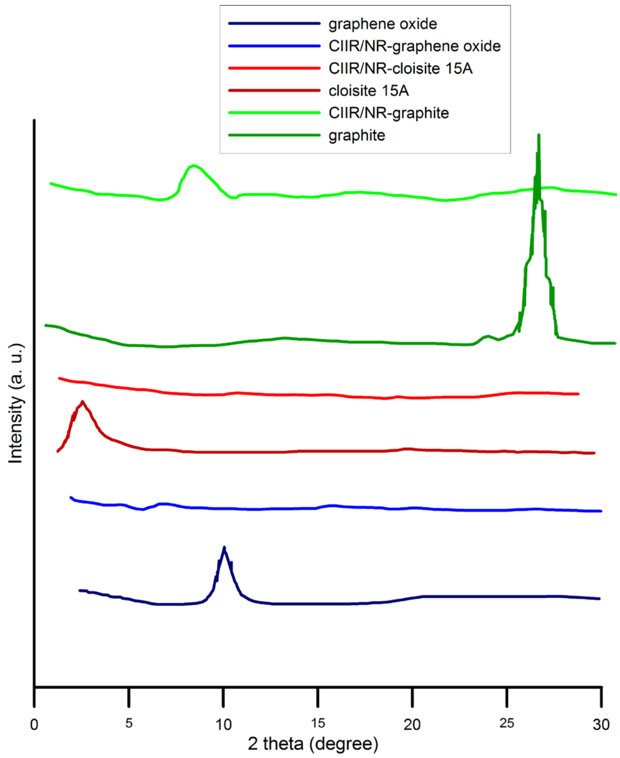

Figure 2. X-ray diffraction of prepared samples.

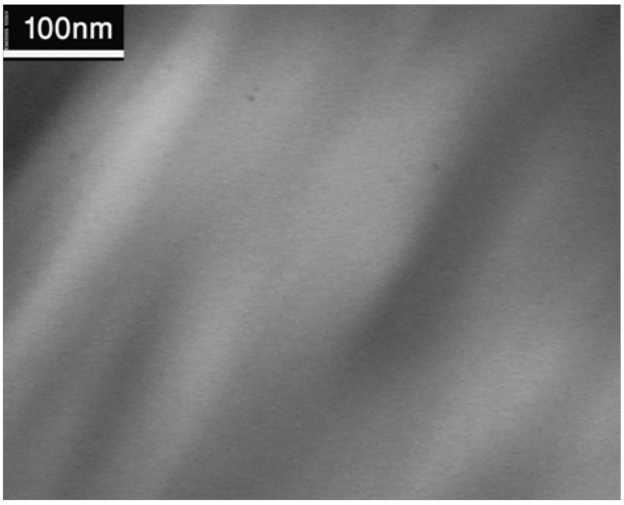

Figure 3. TEM images of CIIR/NA-graphene oxide.

(Figurer 4) also shows the results of the tensile test, as is clear, approximately the reinforcement effect of 4 phr graphite with 1 phr graphene oxide is equal, which is due, firstly, to the proper dispersion (exfoliation) of the nanoparticles of the graphene oxide in the matrix and Secondly, due to the polarization of graphene oxide, it results in greater compatibility between the matrix and the nanofiller compare with graphite.

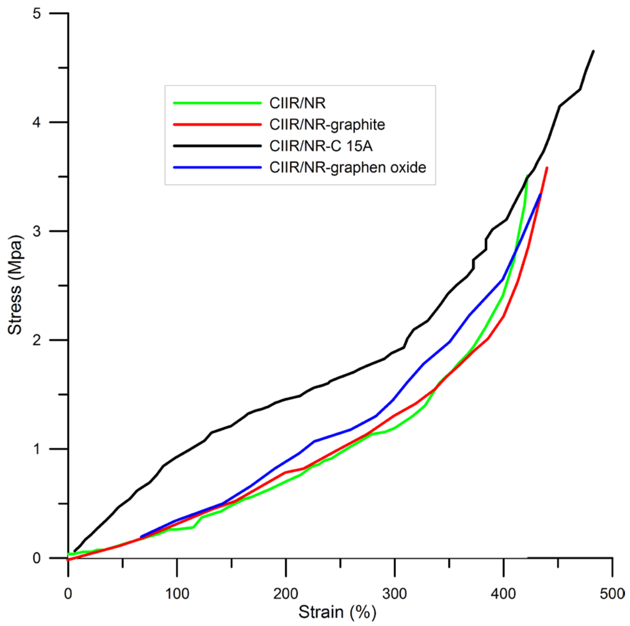

Figure 4. Tensile test results for prepared samples.

4. Conclusion

Enhancing nanoparticles will result in improved properties if they exfoliated in the polymer matrix, and the surface modification of these particles increases matrix-filler compatibility and thus improves dispersion. In the case of graphite and graphene oxide, clearly showed 1 phr of the graphene oxide has more increase properties than 5 phr graphite. The same applies to nanoclay, so that the increased 4 phr modified nanoclay provides high mechanical properties along with proper particle dispersion.

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خواص نانوکامپوزیت‌های پلی‌کلروبوتیل/گرافن اکساید و نانوخاک‌رس از نظر برهمکنش‌های بین پرکننده‌ها

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چکیـــــده

نانوکامپوزیت‌های نانو خاک‌رس اصلاح شده (Cloisite 15A) و گرافن اکساید با لاستیک کلروبوتیل و لاستیک طبیعی SMR20 با موفقیت تهیه شدند. اصلاح سطح نانوذرات باعث بهبود در پراکنش داخل ماتریس لاستیکی شده و خواص قطعه پخت شده مطابق با فرمولاسیون اینرلاینر بهبود قابل توجهی پیدا کرد. ورقه‌ورقه‌شدن ذرات گرافن اکساید و نانو خاک‌رس اصلاح شده توسط آنالیز پراکنش اشعه ایکس و میکروسکوپی الکترونی عبوری (TEM) اثبات گردید، اما در مورد کامپوزیت گرافیت، بین لایه‌ای شدن اتفاق افتاد. مطالعه خواص فیزیکی حاصل از این نانوذرات نشان داد کامپوزیت‌های حاوی نانوذرات اصلاح شده خواص بهبود یافته‌ای از خود نشان می‌دهند.

واژگان کلیدی: پلی‌کلروبوتیل، گرافیت، گرافن اکساید، خاک‌رس نانو و نانوکامپوزیت