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Material properties

Remarkable improvement of failure strain of preferentially aligned short pineapple leaf fiber reinforced nitrile rubber composites with silica hybridization

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Preferentially aligned short fiber reinforced nitrile rubber (NBR) composites with very high moduli at low elongation and high elongation at break were developed by using short and fine pineapple leaf fiber (PALF) and silica as the hybrid (two component) reinforcement. The amount of PALF was fixed at 10 parts (by weight) per hundred of rubber (phr) while that of silica was varied from 0 to 30 phr. Uniaxial NBR composites were prepared and tested for their mechanical properties in the directions both parallel and perpendicular to the fiber axis. Comparison was made against silica-NBR composites of the same total filler loadings. All composites with PALF display very distinct stress-strain curves. The stress rises sharply when the composite is stretched, while that of silica filled composites with the same loading rises gradually. The addition of silica initially lowers the early part of the stress-strain curve but prolongs breaking to greater strains. Further addition of silica raises the early part of the stress-strain curve back to and above that of the lower silica contents. It also significantly increases the elongation at break. Observation of other properties is also reported. Considering all the properties evaluated, reinforcement of NBR with PALF-silica hybrid shows great promise for engineering applications.

1. Introduction

With ever increasing awareness of global warming, environmental problems and resource and oil depletions, much attention is being paid to replacing conventional materials with those from renewable sources. Natural fiber as a reinforcing agent is a main focus due to the excellent mechanical properties that it enables. It has been widely researched and has already reached commercial production for plastic reinforcement. However, despite many studies [1–6], with the exception of cotton, natural fibers have found limited applications in rubbers. In rubbers, fibers provide uniquely characteristic reinforcement, i.e. a sharp rise in stress at relatively low strains. This is not obtainable from rubbers with particulate fillers. This characteristic is important in applications that require highly anisotropic properties but in which large deformation is undesirable.

Natural fibers have a broad distribution of relatively large diameters which range, for example, up to 200 μm for

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jute and 450–500 μm for coir and oil palm. This is thought to be the governing factor that limits the achievable tensile strength, since the size of the fiber is much larger than the size of failure initiation sites in rubbers [7–8]. Such premature failure is often associated with the very low elongation at break of rubbers filled with both natural and synthetic fibers [1,9–12], and this is likely to limit the use of the materials. Therefore, natural fiber reinforced rubbers will be more useful if the elongation at break can be increased.

Hybrid composites, in which two or more types of agent are used to reinforce a common matrix, have been developed to achieve the combined advantages of the constituent materials [13]. Such combinations include using glass fiber with carbon fiber to reduce cost [14], or using the glass fiber to improve compressive strength while retaining the failure strain of carbon fiber composites [15]. There are several other combinations such as cellulose-silica [16] and natural fiber-synthetic fiber [17].

In this work, the known advantages of improving the tensile and tear strengths of rubber by using silica filler [18] were combined with the advantages of short fiber reinforcement. This is the continuation of our recent publication in which pineapple leaf fiber (PALF) was used as the sole reinforcement fiber for nitrile rubber (NBR) [12]. Here are reported the effects of silica content on the mechanical properties of 10 phr PALF-NBR rubber composites.

2. Experimentals

2.1. Materials

PALF: Pineapple leaves from cultivation areas in Kok Kwai District, Amphor Ban Rai, Uthai Thani Province, Thailand were used. PALF was separated by a milling technique developed in our laboratory [19]. The leaves were cut across the long axis into 6 mm long pieces. These were then ground with a disc milling machine in which the soft tissue and fiber bundles were crushed into paste. The paste was then dried and PALF was separated by sieving.

NBR: The rubber was JSR N230SL produced by JSR Corporation Japan. It is medium-high percentage of acrylonitrile (35% acrylonitrile) material with Mooney Viscosity (ML1 + 4(100°C)) of 42.

Rubber chemicals: Synthetic precipitated silica (Tokusil 233), manufactured by OSC Siam silica Co., Ltd Thailand, was used. Resorcinol (Cohedur RS) and hexamethoxymethyl-melamine ether (HMMM) (Cohedur A200) were manufactured by Lanxess Deutschland GmbH, Germany and obtained from Cornell Bros Co,. Ltd, Thailand. All other ingredients were commercial grade chemicals supplied by Rubber Technology Research Center, Mahidol University, Thailand.

2.2. Composite preparation

NBR, PALF and other ingredients, including bonding agent, were mixed on a small two-roll mill. In the mixing process, the nip gap, roller speed and mixing time were kept the same for all formulations. The sequence of ingredient addition is shown in Table 1. Total mixing time was 21 min. Formulations of all mixtures are shown in Table 2. After the mixing was completed, the mixture was taken out using a narrow nip to align the fiber in the machine direction [20]. The compounds were vulcanized at 150°C in a mold of 1 mm thickness using the optimum cure time, T_{90}, determined from Moving Die Rheometry (MDR) results. Care had to be taken in placing the compound sheet in the mold in order to maintain the fiber alignment.

Table 1
Sequence of ingredient addition in compound preparation.

Step	Ingredient	Time (min)
1	NBR	0–1
2	Sulfur	1–2
3	ZnO and Stearic acid	2–5
4	Silica + PALF	5–16
5	Resorcinol and HMMM (if needed)	16–19
6	CBS and TMTD	19–21
Total time		21

2.3. Characterization

Curing behaviour was studied with a Moving Die Rheometer (Rheo TECH MD+, Alpha Technologies, Akron, USA) at 150 °C (ISO 6502).

Tensile properties and tear strength were measured according to ISO 37 and ISO 34, respectively. Specimens were prepared with longitudinal and transverse fiber orientations. Tests were carried out on a universal testing machine (Instron 5569, High Wycombe, UK) with an extensometer attached. A crosshead speed of 500 mm/min and a 1 kN load cell were used. Tensile strength, moduli (or stresses) at 25% and 100% strains, elongation at break and tear strength were determined. The mean value of at least 5 specimens is reported for each property.

Swelling measurements were carried out using rectangular specimens cut from vulcanized sheet. Weighed rectangular specimens were soaked in acetone at room temperature for 72 hours before their weights were determined again. The swelling ratio is the percentage weight gain. Abrasion loss was determined according to ISO 4649 where the cylindrical sample is simultaneously rotating around its axis and sliding over a rotary drum abrader (Zwick Model 6102). For swelling and abrasion loss, the mean value of 3 specimens is reported.

The fracture surfaces of the composites were observed with a scanning electron microscope (SEM) (Hitachi Tabletop Microscope; model TM 1000, Japan).

3. Results

Before considering the general characteristics of PALF/silica-NBR compounds and composites it is worth mentioning that the formulations used are based on a previous study in our laboratory [12] with slight modification. PALF was fixed at 10 phr and, since PALF was found to work best with a bonding agent, all formulations contain a fixed amount of such a compound. Where appropriate to gain better understanding, additional compounds were prepared, tested and compared.