# Studies on Untreated and Silane Treated Ground Granulated Blast Furnace Slag Incorporated Natural Rubber Vulcanizates

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**ABSTRACT**

*A series of natural rubber (NR) vulcanizates have been prepared by partially replacing the carbon black by ground granulated blast furnace slag (GBS). The effect of GBS content on the physico-mechanical properties and cure characteristics of NR composites have been investigated. The effect of with and without silane (Si-69)-modified GBS content on the mechanical behaviors of the NR systems has been reported. The physico-mechanical properties of the rubber vulcanizates were determined before and after heat aging at 90 °C for 72 h. It was observed that GBS filled NR composites exhibits increase in hardness as the GBS filler content increases. Electrical properties of the NR/GBS vulcanizates were also studied.*

*Keywords: NR, GBS, Silane Coupling Agent, Mechanical Behavior, Ageing, Electrical Properties*

## 1. INTRODUCTION

Natural rubber (NR) is an elastomer has been extensively studied because of its wide usage in tyres and O-rings, due to its excellent tensile strength, damping properties and elasticity [1]. Since no elastomer has all the characteristics required in many application areas, elastomers are commonly blended or compounded to improve or modify their properties. NR exhibits outstanding properties such as green strength and tensile strength because it can crystallize spontaneously when it is strained. However, some properties of NR such as modulus, hardness and abrasion resistance need to be improved for some specific applications. Rubbers in their raw state are not good enough for any useful application, except in their vulcanized state. The desired property improvements can be achieved through vulcanization include hardness, tensile strength, modulus and permanent set [2]. These vulcanizate properties can be optimized when rubbers are compounded with suitable property-enhancing additives. Among these additives, zinc oxide (ZnO) in conjunction with stearic acid acts as an activator (or activating system) during sulphur vulcanization [3]. Sulphur vulcanization alone is a very slow process; hence, the essence of activating the vulcanization process is to increase the efficiency of crosslink formation and consolidation of crosslink density [4]. The object of compounding is to select the most suitable combination of additives in their correct proportion in order to enhance the vulcanization or processing characteristics and/or

the end use performance of the vulcanizate [5]. The effects of different types of fillers such as carbon black, silica, calcium carbonate, etc., on NR compounds have been studied, in search of improvements on its physico-mechanical properties. Recently, the application of fillers of organic nature has attracted interest due to their low cost, renewable and environmental friendly nature. Studies have reported on the modulus enhancement of rubbers by incorporating natural materials, for example; egg shell powder[6], cherry seed shell [7], china clay and $CaCO_3$ [8-10] and ground rice husk [11-12]. It is known that in the case of filled vulcanizates, the efficiency of reinforcement depends on a complex interaction of several filler related parameters. These include filler size, filler shape, filler dispersion, surface area, surface reactivity, filler structure and interfacial adhesion between fillers and rubber matrix [13].

Scientists now focus on the use of natural materials for the development of composite materials. The attention is due to their renewable characteristics. To improve on the mechanical properties and obtain the desired characteristics that will be suitable for particular application different matrices are used for reinforcing natural fillers at various particle sizes. Fillers having cellulose, hemicellulose and lignin are being investigated for the suitability of replacing the synthetic fibres/fillers. The use of these natural fillers has been due to its economic advantage during

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processing, high specific strength, relatively low density and the biodegradability, thereby reducing environmental pollution [14-17]. Zaini et al. [18] reported that for all filler sizes, the mechanical properties of polypropylene (PP)/kaolin composites decreased with increasing filler content. The incorporation of fly ash into PP matrix resulted into better tensile modulus, while the tensile strength, elongation at break and impact strength decreased with increasing filler loading [19-21]. A relationship between decreasing particles size and increasing impact strength in mineral-filled PP was also reported [22].

The aim of this contribution is to assess the potential utilization of ground granulated blast furnace slag (GBS) in NR compounds. GBS, an absolutely low cost inorganic waste product continues to pose a hazard and must thus be used in applications that help to curb environmental pollution. The main constituent of GBS is $CaCO_3$ and silica. NR filled GBS vulcanizates prepared by the conventional method, with and without the treatment of coupling agent are used for comparison, keeping the recipe same. Silane coupling agent was selected as it is used to improve the reinforcing capability of the fillers [23]. Silane coupling agent is recognized as an efficient coupling agent in the rubber field [24-25]

## 2. EXPERIMENTAL
### 2.1. Materials
Natural rubber (NR) of grade ISNR 20 was obtained from the Rubber Board, Kottayam, Kerala. Ground granulated blast furnace slag (GBS) was obtained from Speciality Minerals, Baroda, India. GBS is prepared by quenching molten iron blast furnace slag in water or steam, to produce a glassy granular product, then dried and ground into a fine powder. The general chemical composition of GBS is; 40% CaO, 35% $Si_2O_3$, 13% $Al_2O_3$ and 8% MgO. The GBS average particle size is 1.5 µm and specific gravity is 2.91. It was dried before compounding. The coupling agent [(Si-69): bis(3-triethoxy silyl propyl)tetrasulfide] was purchased from Degussa-Huls, Germany.

### 2.2. Silane treatment of GBS
The silane coupling agent (1 g) was mixed [26-27] with ethyl alcohol (100 ml) to make a solution for applying on to the filler (100 g). The filler was mixed with the solution of coupling agent with stirring to ensure uniform distribution of the coupling agent; mixing was continued for 30 min. The silane treated GBS filler was then dried at 80 °C in a hot air oven for about 3 h for complete evaporation of the ethanol.

### 2.3. Compounding of NR with fillers

Table 1 shows the formulations and materials used in this research study. Rubber was first masticated and known amount of fillers with other ingredients were mechanically mixed and compounded on a two roll mill. Six compositions of NR/GBS and five compositions of NR/silane treated GBS were prepared respectively in identical conditions. The films were scratched away from the rollers and then compression molded as thick uniform sheets. Specimens of desired size were cut from this sheet for mechanical, physical and electrical testing.

### 2.4. Techniques
Cure characteristics of NR/GBS formulations were studied using a Moving Die Rheometer 2000 (MDR 2000, USA) at 160 ℃ according to ASTM D 2528. Tensile properties of the vulcanized NR sheets were determined using universal testing machine (ZWICK Z 2.5, Roell, Germany) at room temperature (25±2 °C), at a uniform speed of separation of 500 mm/min according to ASTM D 412 method. Compression set testing was done as per ASTM D395-B method and testing conditions was 100 ℃ for 24 h. Hardness test was carried out using a shore A Durometer in accordance to ASTM D2240. Ageing was carried out in an air-circulating oven at 90 ℃ for 72 h. Different electrical properties like volume resistivity, surface resistivity, capacitance, dielectric loss, permittivity and breakdown strength were monitored as per ASTM 257. The AC conductance and capacitance of sample were measured with an LCR meter (HP4263B). The specimen, with a thickness of 2 mm and size of 100 x 100 mm was placed between the guarded (top) and unguarded (bottom) electrodes. The readings were recorded at the end of 60 sec.

## 3. RESULTS AND DISCUSSION
Table 2 shows the cure characteristics of the untreated and silane treated GBS fillers loaded NR formulations. It can be seen that the minimum (ML) and maximum (MH) torque decreases slightly from 6.43 to 5.32 and 64.49-59.53 dNm respectively with increasing filler loading. The addition of filler decreased the maximum torque value which indicated that the addition of GBS does not help better filler-matrix interaction. The decrease in minimum torque with GBS loading was mainly attributed by filler-filler interaction rather than the rubber filler interaction. These results were partially attributed to the fact that GBS is rigid filler. On the other hand, the introduction of GBS filler showed a shorter optimum curing time (Ts90) curing time upto 10 phr (parts per hundred parts of rubber) (1.56-1.51 min), indicating that GBS facilitated the curing process for rubber composites. At lower GBS content the presence of crosslinked precursors decreased the cure time and as the concentration of GBS increased, the

agglomeration of GBS might retard the rate of vulcanization of NR formulations. The scorch time (Ts2) of filled composites took on increasing trend from 0.57 to 0.71 with an increase of GBS content. Silane treated GBS filled NR formulations always showed a higher cure characteristic values (ML and MH) as compared to untreated filler loaded NR formulations (Table 2). As torque values are related to viscosity and processability of the compounds, the processing of compounds becomes easier with increasing amount of silane treated fillers. The decrease in optimum cure time (Ts90) from 1.48 to 1.45 upto10 phr loading indicates an interference effect on the crosslink formation of the rubber caused by the presence of the Si69. This is also supported by the change in scorch time (Ts2) for the NR with silane treated GBS formulations.

### 3.1 Physico-mechanical properties

The results of the physico-mechanical properties of NR vulcanizates filled with modified and unmodified GBS is addressed in Table 3.

### (a) Surface hardness

Dependence of the hardness on the amount of GBS filler in NR composites is given in Table 3. The obtained surface hardness of GBS loaded NR composites lies in the range 53-57 IRHD. It is seen that, hardness of the NR/GBS composites marginally increases with increase in filler content. This was simply due to the fact that the incorporation of more rigid filler particles into rubber, reduced the elasticity of the rubber chains and led to more rigid rubber materials. It is observed from Table 3, that the silane treated GBS loaded NR composites have slightly higher values of surface hardness than the untreated filler loaded NR vulcanizates. The surface hardness of silane treated GBS filler loaded NR formulations lies in the range 56-60 IRHD. The higher hardness of the silane treated GBS loaded NR composites may be attributed to the additional filler-polymer interaction associated with the aid of coupling agent [28].

### (b) Tensile behavior

The effect of filler loading on the tensile strength of the untreated and silane treated NR/GBS composites is given in Table 3. There was a slight improvement in the tensile strength upto 5 phr of GBS loading. The experimental results showed that the addition of untreated filler above 5 phr slightly reduced the tensile strength of the NR/GBScomposites. The reduction in tensile strength above 5phr for the GBS filler filled NR composites may be attributed to the poor interaction between rubber and filler particles [29]. NR is hydrophobic by nature, while GBS is hydrophilic. Hence, these two are incompatible constituents, which led to weaken the interfacial

interactions between theconstituents. Supaphol et al [30] made a similar observation for for CaCO3-filled syndiotactic polypropylene (PP) composites. The effect of silane coupling agent coated GBS filler filled NR composites on tensile behavior can also be seen in Table 3. A trend of decreasing tensile strength from 7.38 to 6.42 MPa with filler loading upto 15 phr was observed. The presence of the coupling agent had improved the interfacial interaction between the matrix and filler, which in turn increases the tensile strength of the treated composites as given in Table 3. According to Krysztafkiewicz and Domka [31], the use of silane permits, an increase in strength.Elongation at break of the NR/CB (410%) was higher than the corresponding values for the treated or untreated filler loaded NR vulcanizates at any concentration of MK filler. When 5 phr of the fillers were added to the composites, the elongation at break showed a drop to 390 %. A marked reduction in percentage elongation at break from 410 to 365 with increase in GBS content from 0 to 15 phr in NR systems was noticed (Table 3). When the percentage of filler loading was increased, the ductility of the NR/GBS composites was greatly decreased. This demonstrates that the fillers had hardened the composites and reduced their ductility. From Table 3, after incorporating silane treated GBS there was further decrease in the percentage elongation at break which means that the Table 3 shows Young's modulus for the untreated and silane treated NR/GBS composites. Fillers are known to increase the modulus, provided that the modulus of the filler is higher than that of the polymer matrix.

Luyt et al [32] reported that the distribution of filler particles in a polymer matrix greatly influences the modulus of the composites. Young's modulus of the composites slightly increased from 4.38 to 4.45 MPa in untreated and from 4.38 to 4.48 MPa in treated NR/GBS composites as the filler loading increased from 0 phr to 5 phr. The presence of the fillers upto a certain extent had reduced the ductility of the composites and increased their stiffness. This result agrees with those from George et al [33], where the LDPE was reinforced with pineapple-leaf fibers. However, further increase in filler loading, marginally reduces the modulus. It was observed that modulus of composites slightly decreases with increase in filler content. This was rather not surprising, as fillers are customarily added to enhance the modulus. Generally, the presence of load bearing or dissipating microelements with their phase boundaries leads to a decrease in micro stress concentration. At higher volume fraction of filler stresses tend to initiate local rupture at micro domain boundaries. Thus, cracks are limited by micro in homogeneity and cannot proceed very far without encountering modulus changes diverting their path and reducing

**Table 1**. Typical formulations of NR/GBS vulcanizates.

| Formulation, phr | 1 | 2 | 3 | 4 | 5 | 6 |
|---|---|---|---|---|---|---|
| NR | 100 | 100 | 100 | 100 | 100 | 100 |
| Zinc oxide (ZnO) | 4.0 | 4.0 | 4.0 | 4.0 | 4.0 | 4.0 |
| Stearic acid | 2.5 | 2.5 | 2.5 | 2.5 | 2.5 | 2.5 |
| Oil | 14 | 14 | 14 | 14 | 14 | 14 |
| Antioxidant | 1.0 | 1.0 | 1.0 | 1.0 | 1.0 | 1.0 |
| Calcium carbonate | 30 | 30 | 30 | 30 | 30 | 30 |
| Carbon black | 30 | 25 | 25 | 25 | 25 | 25 |
| Paraffin wax | 7 | 7 | 7 | 7 | 7 | 7 |
| Dibenzothiazole disulphide (MBTS) | 1.0 | 1.0 | 1.0 | 1.0 | 1.0 | 1.0 |
| TMTD | 0.2 | 0.2 | 0.2 | 0.2 | 0.2 | 0.2 |
| Sulphur | 2.5 | 2.5 | 2.5 | 2.5 | 2.5 | 2.5 |
| PVI (retarder) | 0.2 | 0.2 | 0.2 | 0.2 | 0.2 | 0.2 |
| GBS | 0 | 5 | 7.5 | 10 | 12.5 | 15 |
| Sample code | GBS | GBS5 | GBS7.5 | GBS10 | GBS12.5 | GBS15 |

For silane treated filler code number is; GBST5, GBST 7.5, GBST10, GBST12.5 and GBST15 respectively.

**Table 2**. Cure characteristics of untreated GBS loaded NR formulation.

| Properties | Ground granulated blast furnace slag (phr) | | | | | |
|---|---|---|---|---|---|---|
| | 0 | 5 | 7.5 | 10 | 12.5 | 15 |
| **Untreated GBS** | | | | | | |
| Scorch time (Ts2, min) | 0.57 | 0.62 | 0.63 | 0.64 | 0.69 | 0.71 |
| Optimum cure time (Ts90, min) | 1.56 | 1.55 | 1.53 | 1.51 | 1.67 | 1.64 |
| Minimum torque (ML, dNm) | 6.43 | 6.40 | 6.36 | 6.06 | 5.59 | 5.32 |
| Maximum torque (MH, dNm) | 64.49 | 63.73 | 63.12 | 63.00 | 62.51 | 59.53 |
| **Silane treated GBS** | | | | | | |
| Scorchtime(Ts2,min) | | 0.72 | 0.71 | 0.71 | 0.71 | 0.74 |
| Optimum cure time (Ts90, min) | | 1.48 | 1.43 | 1.45 | 1.65 | 1.62 |
| Minimum torque (ML, dNm) | | 6.63 | 6.74 | 6.71 | 6.54 | 5.99 |
| Maximum torque (MH, dNm) | | 65.04 | 65.49 | 65.53 | 63.86 | 63.06 |

their energy. A significant reduction in tensile modulus with increase in filler content above 7.5 phr is expected as GBS doesn't have strongpolymer-filler interactions, which don't restrict the mobility of NR network. In all the cases silane treated GBS shows slightly better properties as compared to untreated GBS filler loaded NR systems.

### (c) Compression set properties

The effect of GBS content on the compression set behavior of NR compounds has been studied and the obtained experimental results are given in Table 3. The table showed the variation of the percentage of compression set of GBS loaded NR compounds. The compression set increases from 48.4 to 54.3% with increase in the GBS filler content upto 7.5 phr [34]. From Table 3, it is also noticed that the compression set increases from 54.4 to 56.1% with the increase in silane treated GBS filler content upto 7.5 phr. This is due to a better reinforcement between the filler and the elastomer after treating GBS with silane.

### (d) Effect of heat ageing on tensile behaviors

The industrially important aspects of aging are the changes in physical properties such as the tensile strength, the hardness, or the modulus of elasticity. If the conditions are too severe, the rubber may rapidly become unserviceable. The aging process of NR is complex, but it is known that oxidation is a significant degradation process. The rate of degradation is significantly accelerated at higher temperatures. The measured mechanical properties of thermally aged NR composites are given in Table 4. However, at 90°C of aging, the values of modulus at 200 % elongation increased. Ahagon et al. [35] and Baldwin et al. [36], in their studies of accelerated aging of rubber compound, they observed that the modulus increase and later reduction, depending on aging mechanism. The effect of aging temperature on modulus is due to the complexity of reactions taking place in the cured rubber compound. This phenomenon is a post-curing effect which tends to increase when aging temperature increases. Clarke et al. [37], in

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**Table 3.** Effect of untreated and silane treated GBS content on the physico-mechanical properties of NR/GBS composites.

| Sample code | Tensile strength (MPa) | Elongation at break (%) | Tensile modulus (MPa) @ 200% | Surface hardness (IRHD) |
|---|---|---|---|---|
| **Untreated GBS** | | | | |
| GBS0 | 6.72 | 355 | 4.57 | 57 |
| GBS5 | 6.75 | 310 | 4.63 | 58 |
| GBS7.5 | 6.69 | 303 | 4.61 | 59 |
| GBS10 | 6.27 | 265 | 4.52 | 60 |
| GBS12.5 | 6.17 | 271 | 4.38 | 61 |
| GBS15 | 6.01 | 275 | 4.23 | 62 |
| **Silane treated GBS** | | | | |
| GBST5 | 6.89 | 289 | 4.71 | 60 |
| GBST7.5 | 6.83 | 276 | 4.68 | 61 |
| GBST10 | 6.48 | 235 | 4.59 | 61 |
| GBST12.5 | 6.25 | 254 | 4.40 | 62 |
| GBST15 | 6.13 | 256 | 4.25 | 63 |

**Table 4.** Effect of heat ageing on untreated and silane treated GBS content on the physico- mechanical properties of NR/GBS composites.

| Sample code | Tensile strength (MPa) | Elongation at break (%) | Tensile modulus (MPa) @ 200% | Surface hardness (IRHD) |
|---|---|---|---|---|
| **Untreated GBS** | | | | |
| GBS0 | 6.72 | 355 | 4.57 | 57 |
| GBS5 | 6.75 | 310 | 4.63 | 58 |
| GBS7.5 | 6.69 | 303 | 4.61 | 59 |
| GBS10 | 6.27 | 265 | 4.52 | 60 |
| GBS12.5 | 6.17 | 271 | 4.38 | 61 |
| GBS15 | 6.01 | 275 | 4.23 | 62 |
| **Silane treated GBS** | | | | |
| GBST5 | 6.89 | 289 | 4.71 | 60 |
| GBST7.5 | 6.83 | 276 | 4.68 | 61 |
| GBST10 | 6.48 | 235 | 4.59 | 61 |
| GBST12.5 | 6.25 | 254 | 4.40 | 62 |
| GBST15 | 6.13 | 256 | 4.25 | 63 |

**Table 5.** Change in weight of NR/GBS (untreated and treated) after exposure to different chemical reagents for 7 days.

| Sample Code | % Change in weight for 7 days at room temp. for various chemical reagents | | | | | |
|---|---|---|---|---|---|---|
| | HCl(10%) | FAS(10%) | KMnO$_4$(10%) | Benzene | NaOH(10%) | H$_2$O |
| **Untreated GBS** | | | | | | |
| GBS0 | 1.10 | 0.79 | D | 139 | 0.47 | 2.54 |
| GBS5 | 1.34 | 0.66 | D | 133 | 0.46 | 2.48 |
| GBS10 | 1.42 | 0.67 | D | 137 | 0.51 | 2.07 |
| GBS15 | 1.45 | 0.44 | D | 139 | 0.65 | 2.84 |
| **Silane treated GBS** | | | | | | |
| GFST5 | 0.88 | 0.57 | D | 131 | 0.59 | 1.82 |
| GBST10 | 0.67 | 0.43 | D | 129 | 0.60 | 1.42 |
| GBST15 | 0.49 | 0.49 | D | 135 | 0.62 | 1.01 |

their study on aging effect on tensile strength of NR compound, also show that cross-linking reaction increase with increasing aging temperature. On ageing a noticeable increase in the hardness is observed. Silane treatment on GBS makes it rigid thereby it increases the surface hardness. A reduction in the tensile strength and elongation at break was noticed after heat ageing. The accelerated aging of rubbers that contain unsaturated bonds normally results in a reduction in their strength properties.

### 3.2 Chemical resistance

The resistance of untreated and treated NR/GBS composites to various chemical reagents was measured by immersing the specimens in different chemical reagents/solvents. In the present investigation various chemical reagents such as acid, alkali, oxidizing agent and water were chosen and the chemical resistance of NR/GBS composites were investigated. The measured change in

**Table 6.** Volume and surface resistivity of untreated and treated NR/GBS composites.

| Sample code | Volume resistivity in $10^8$ $\Omega$-cm | Surface resistivity in $10^9$ $\Omega$-cm |
|---|---|---|
| **Untreated GBS** | | |
| GBS0 | 4.74 | 83.54 |
| GBS5 | 32.04 | 105.3 |
| GBS7.5 | 37.02 | 126.2 |
| GBS10 | 38.55 | 132.2 |
| GBS12.5 | 39.98 | 133.9 |
| GBS15 | 42.96 | 138.7 |
| **Silane treated GBS** | | |
| GBST5 | 31.01 | 102.6 |
| GBST7.5 | 34.07 | 110.9 |
| GBST10 | 37.73 | 128.0 |
| GBST12.5 | 38.80 | 131.6 |
| GBST15 | 40.85 | 135.3 |

**D\*** denotes disintegrated in KMnO4.

percentage weight of NR/GBS composites after exposure to different chemical reagents is tabulated in Table 5. From the table it was noticed that, there was no significant change in weight for all the chemical reagents used under this investigation except for benzene. From this study it was noticed that, GBS filled NR vulcanizates are very sensitive to organic solvents as compared to other reagents used and the resistance to benzene decreased with increase in GBS content. This conclusion was drawn on the basis of percentage change in weight of the composite specimens. The samples degraded in KMnO$_4$ and it indicates that NR vulcanizates are very sensitive to the oxidation media. From the study it was also observed that, there was no significant change in surface color of the specimens. From the table it is also experimented

that the silane treated GBS is more resistant to the chemical environments.

### 3.3 Electrical properties
#### (a) Volume and Surface Resistivity
As seen from the Table 6, both the volume and surface resistivity of the composites increase with increase in untreated and treated GBS filler content. The increase in resistivity may be due to the enhancement in interfacial interaction between carbon black and GBS aggregates and reduction in polarity in the composites during vulcanization and also due to the introduction of high resistivity GBS. The silane treated GBS filled NR vuilcanizates have lower volume and surface resistivity than the untreated filler loaded systems. The reduction in electrical resistivity due to the presence of silicate group, where silicate consists of silicon with oxygen as the ligand silicate anions, with a negative net electrical charge [38].

#### (b) Dielectric loss and break down voltage
The dielectric loss or tan delta values indicate the bulk dielectric losses in the insulating material due to the conduction processes in the dielectric that cause the flow of leakage current. For any electrical insulation system, a low dielectric loss value is always desired when the material is to be used as a dielectric media. Higher losses in an insulating material cause heating of the product made of this material and may even lead to thermal breakdown. A significant reduction in the dielectric loss values for untreated and treated GBS filler filled NR vulcanizates was noticed (Table 7). One of the basic important parameters of an insulating material is its breakdown strength. Breakdown in solid insulating materials may result from both electric and thermal processes that occur under the action of the electric field. From Table 7, it can be observed that the electrical breakdown strength and voltage increased with increase in the concentration of both untreated and silane treated GBS filler.

#### (c) Capacitance and Permittivity
Table 7 shows a drastic increase in the capacitance values of the NR vulcanizates with increase in untreated filler concentrations and it lies in the range 228-480 pF. However, the variation in capacitance values is not systematic with the composition of the silane treated filler filled NR formulations. As GBS content increases the agglomeration will be more which leads to an increase in the capacitance. As seen from Table 7, permittivity values increased with increase in the GBS content. But addition of silane treated GBS a decrease in the permittivity values is observed. The permittivity is associated with the mobility of polymer chains. The lower permittivity of silane treated NR/GBS composites is due to a better interfacial adhesion between filler and matrix.

**Table 7**. Capacitance, permittivity anddielectric properties of NR/GBS composites.

| Sample code | Capacitance (pF) | Permittivity (pF) | Dielectric loss (Tan δ) | Breakdown voltage in  kV | Breakdown strength in kV/mm |
|---|---|---|---|---|---|
| **Untreated GBS** | | | | | |
| GBS0 | 228 | 67.4 | 0.46 | 9.6 | 3.39 |
| GBS5 | 296 | 65.4 | 0.42 | 9.9 | 3.43 |
| GBS7.5 | 319 | 68.9 | 0.35 | 10.3 | 3.55 |
| GBS10 | 353 | 72.2 | 0.34 | 10.5 | 3.61 |
| GBS12.5 | 428 | 80.1 | 0.25 | 10.8 | 3.89 |
| GBS15 | 480 | 91.3 | 0.19 | 10.9 | 4.14 |
| **Silane treated GBS** | | | | | |
| GBST5 | 679 | 66.9 | 0.40 | 10.2 | 3.69 |
| GBST7.5 | 518 | 74.1 | 0.33 | 10.6 | 3.75 |
| GBST10 | 704 | 73.5 | 0.32 | 11.2 | 3.83 |
| GBST12.5 | 593 | 82.4 | 0.24 | 12.1 | 3.92 |
| GBST15 | 618 | 92.1 | 0.18 | 12.5 | 4.29 |

## 4. CONCLUSIONS

From the current investigation of physico-mechanical properties and electrical properties of NR/GBS vulcanizates the following conclusions were derived:

i). The maximum value of tensile strength was achieved at 5 phr for untreated and silane coupling agent treated GBS filled NR vulcanizates.

ii). A significant improvement in tensile modulus and surface hardness of the composites after heat ageing was noticed. A slight reduction in tensile strength and marked reduction in percentage elongation at break after heat ageing were noticed.

iii). Volume and surface resisitivity values increased with increase in filler laoding.

iv). A significant reduction in dielectric and an increase in capacitance, permittivity and breakdown strength were noticed with the increase in both silane treated and untread GBS content in the vulcanizates.

From the above observations it was inferred that 5 phr of GBS filled NR vulcanize is the optimized formulation.

## 5. REFERENCES

[1]. S. Riyajan, J.T. Sakdapipanich (2009) Investigation of the preparation and physical properties of a novel semi-interpenetrating polymer network based on epoxised NR and PVA using maleic acid as the crosslinking agent, *Chemical Engineering Journal*, **153**: 199-205.

[2]. A.T. Koshy, B. Kuriakose, S.Thomas (1992) Studies on the effect of blend ratio and cure system on the degradation of natural rubber-ethylene-vinyl acetate rubber blends, *Polym. Degradation and Stability*, **36**: 137-147.

[3]. F. Findik, R. Yilmaz, T. Koksal (2004) Investigation of mechanical and physical properties of several industrial rubbers, *Materials & Design*, **25**: 269-360 .

[4]. M.T. Ramesan (2004) The effects of filler content on cure and mechanical properties of dichlorocarbene-modified styrene-butadiene rubber/carbon black composites, Journal of Polymer Research, 11, 333-348.

[5]. T. Kurian, P.P. De, D. Khastgir, D.K. Tripathy, S.K. De, D.G. Peiffer (1995) Reinforcement of EPDM-based ionic thermoplastic elastomer by carbon black, *Polymer,* **36:** 3875-3884.

[6]. R. Jayasankar, N. Mahindran, R. Ilangovan (2010) Studies on concrete using fly ash, rice husk ash and egg shell powder, *International Journal of Civil and Structural Engineering*, **1**: 362-372.

[7]. E. Osabohienn, S.H.O. Egboh (2007) Cure characteristics and physic-mechanical properties of carbonized bamboo fibre filled natural rubber vulcanizates, *Journal of Applied Sciences and Environmental Management*, **11**: 43-48.

[8]. M.A. Bello (2001) Polymers-the chemistry and technology of modern materials, Concept publisher Ltd., Lagos, Nigeria, 227-228.

[9]. C.M. Blow, C. Hepburn (Ed) (1982) "Rubber Tech. and Manufacture", Butterworth Scientific, London, 2nded, 1-540.

[10]. A. Whelan, K.S. Lee (1979) "Development in Rubber", Applied Science Publisher Ltd., London, 184.

[11]. S. Salina Sarkawi, Yusof Aziz (2003) Ground rice husk as filler in rubber compounding, Journal Technology, University Technology Malaysia, 39, 135-148.

[12]. A.R. Ahmed (2004) "Study of the physical properties for unsaturated polyester resin reinforced NR and SBR", M.Sc. Thesis, college of Education, Tikrit University.

[13]. M. Morton (1995) Rubber Technology: Chapman & Hall, 3rd Edition, Harrisonburg, Virginia, USA.

[14]. H.G.B.Premalal, H. Ismail, A.Baharin (2002) Comparison of the mechanical properties of rice husk powder filled polypropylene composites with talc filled polypropylene composites, *Polymer Testing,* **21**, 833-839.

[15]. A.K. Bledzki, J. Gassan (1999) Composites reinforced with cellulose based fibres, *Progress in Polymer Science,* **24**: 221-228.

[16]. N. Rattanasom, T. Saowapark, C. Deeprasertkul (2006) Reinforcement of natural rubber with silica/carbon black hybrid filler, *Polymer Testing*, **26**: 369-377.

[17]. K.G.Satyanarayana, A G.G.C.Rizaga, F.Wypych (2009) Biodegradable composites based on lignocellulosic fibers-an overview, *Progress in Polymer Science,* **34**: 982-1021.

[18]. R.E. Zaini, S.M. Rowell, A.R. Sanadi (1995) Recent developments in annual growth lignocellulosic as reinforcing fillers in thermoplastics, Proceed. of 2nd Biomass Conf. of the Americas: Energy, Environment, Agriculture and Industry, 1171-1180.

[19]. P. Nimityongskul, T. U.Daladar (1995) Use of coconut husk ash, corn cob ash and peanut shell ash as cement replacement, *Journal of Ferrocement*, **25**: 35.

[20]. I. Ahmad, D.R. AbuBakar, S.N. Mokhilas (2005), Recycled PET for Rice Husk/Polyester Composites *Asian Journal for Science and Technology Development*, **22**: 345.

[21]. N.M. Stark, R.E. Rowlands (2003) Effects of wood fiber characteristics on mechanical properties of wood/polypropylene, *Wood and Fiber Science,* **35**: 167-174.

[22]. L. Jiken, G. Malhmmar, R.Selden (1991) The effect of mineral fillers on impact and tensile properties of polypropylene, *Polymer Testing*, **10**: 329-344.

[23]. A.S. Hashim, B. Azahari, Y. Ikeda, S.Kohjiya (1998) The effect of bis(3-tri-ethoxysilyl propyl) tetrasulfide on silica reinforcement of styrene-butadiene rubber, *Rubber Chemistry and Technology*,**7**: 289-299.

[24]. S. Taj, M.A.Munawar, S.Khan (2007) Natural fiber reinforced polymer composites, Proceedings of the *Pakistan Academy of Sciences*, 44: 129-144.

[25]. H.Yan, G.Tyan, K.Sun, Y.Zhiang, Y.Zhiang (2005) Effect of silane coupling agent on the polymer-filler interaction and mechanical properties of silica-filled natural rubber, *Journal of Polymer Science Part B: Polymer Physics*, **43**: 573-584.

[26]. A.S. Hashim, B. Azahari, Y. Ikeda, S. Kohjiya (1998) The effect of bis(3-

triethoxysilylpropyl) tetrasulfide on silica reinforcement of styrene-butadiene, *Rubbe Rubber Chemistry and Technology*, **7:** 289-300.

[27]. R.J. Pickwell (1982) Bis(triethoxysilylethyl tolylene)-polysulfide, a scorch-resistant silane coupling agent for mineral-filled elastomers, *Rubber Chemistry and Technology*, **56:** 94.

[28]. Z.A.M. Ishak, A.A. Bakar (1995) An investigation on the potential of rice husk ash as fillers for epoxidized natural rubber, *Europian Polymer Journal*, **31:** 259-269.

[29]. H.D. Hozman, Z.A.M. Ishak (1998) Rubber wood-high-density polyethylene composites: Effect of filler size and coupling agents on mechanical properties, *Journal of Applied Polymer Science*, **69**: 1993-2004.

[30]. P. Supaphol, W. Harnsir, J.Junkasem (2004) Effects of calcium carbonate and its purity on crystallization and melting behavior, mechanical properties and processability of syndiotactic polypropylene, *Journal of Applied Polymer Science,* **92**: 201-212.

[31]. A. Krysztafkiewicz, L. Domka (1986) Effect of silane coupling in filler rubber vulcanizates, *Plastics and Rubber Processing and Applications*, **6**: 197-203.

[32]. A.S.Luyt, J. A. Molefi, H. Krump (2005) Thermal, mechanical and electrical properties of copper powder filled low-density and linear low-density polyethylene composites, *Polymer Degradation Stability*, **91**: 1629.

[33]. J.George, S.S. Bhagawan, S.Thomas (1997) Electrical of pineapple fiber reinforced polyethylene composites, *Journal of Polymer Engineering* 17: 383-404.

[34]. M.V. Schuur, R.J. Gaymans (2005) Influence of chemical crosslinks on the elastic behavior of segmented block copolymers, *Polymer*, **46**: 6862-6868.

[35]. A. Ahagon, M. Kida, H. Kaidou (1990) Aging of Tire Parts during Service. I. Types of aging in heavy-duty tires, *Rubber Chemistry and Technology,* **63**: 683-697.

[36]. M. Baldwin, D.R. Bauer, K.R. Ellwood (2005) Accelerated aging of tires, Part II, *Rubber Chemistry and Technology*, **78**: 336-353.

[37]. J. Clarke, E.F. Ngolemasango, M. Bennett (2006) Kinetics of the effect of ageing on tensile properties of a natural rubber compound, *Journal of Applied Polymer Science*, **102**: 3732-3740.

[38]. S. Kermit, K.N. Chitra, P. Subramaniam, T.C. Ward (2003) Effect of penetrant size and shape on its transport through a thermoset adhesive: I. n-alkanes, *Polymer*, **44**: 3061-3069.