

## UTILIZATION OF TEA WASTE AS AN ALTERNATIVE FILLER FOR NATURAL RUBBER

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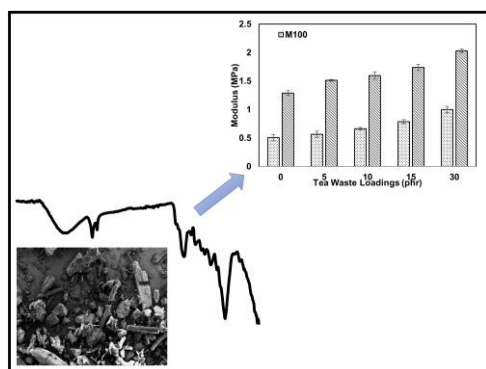
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### Graphical abstract



### Abstract

Tea leave waste (TW) is considered worthless after being discarded. It frequently dumps to the environment without providing the best solution on it. This has come to a final solution to utilize this material as an alternative filler for natural rubber (NR). To optimize the suitable content of TW filled NR, the study was evaluated through the curing characteristics, tensile properties and the morphology of the composites. The results have shown some interesting findings. As for the curing characteristics, the scorch and curing times were decreased with increasing the TW loadings. This is due to the amine group available in TW which fastened the vulcanization process. Harder phase of TW also affected to increase the maximum torque of the compound and this correlated to the tensile modulus of the composites. However, the tensile strength was found to be decreased as the TW loading increased. This could be due to the incompatibility or poor adhesion between TW and matrix of rubber. Considering the amounts of TW, a 10 phr of TW was found to be a content of choice for natural rubber. It provided a balance set of properties regardless whether the mechanical properties and morphological characteristics of the natural rubber.

Keywords: Natural rubber, tea waste, filler, tensile properties, morphology

### Abstrak

Sisa daun teh (TW) adalah dianggap tidak bernilai selepas dibuang. Lazimnya TW dibuang ke persekitaran tanpa penyelesaian yang baik. Dalam kajian ini, TW telah digunakan sebagai pengisi alternatif di dalam getah asli. Untuk mengoptimalkan kandungan TW yang sesuai di dalam getah asli, kajian telah dilakukan untuk menilai ciri-ciri pematangan, sifat tensil dan morfologi getah asli. Keputusan yang diperolehi menunjukkan penemuan yang menarik. Untuk ciri-ciri pematangan, masa skorj dan masa pematangan berkurang dengan penambahan kandungan TW. Ini disebabkan kumpulan amina yang wujud di dalam TW yang mempercepatkan proses pematangan. Fasa TW yang lebih keras juga meningkatkan tork maksimum sebatian getah asli yang menunjukkan korelasi dengan modulus tensil komposit. Walau bagaimanapun kekuatan tensil didapati berkurang dengan penambahan kandungan TW. Ini adalah disebabkan ketidakserasian atau lekatan lemah di antara TW dan matrik getah. Dengan mengambil kira kandungan TW, 10 phr adalah kandungan yang paling sesuai untuk getah asli. Ia menyediakan satu set sifat-sifat yang seimbang dengan mengambil kira sifat mekanikal dan ciri-ciri morfologi getah asli.

Kata kunci: Getah asli, sisa daun teh, pengisi, sifat-sifat mekanik, morfologi

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## 1.0 INTRODUCTION

Rubber products rely greatly on the use of filler in their ingredients. Most fillers are incorporated for many reasons. For examples, to facilitate the processing of rubber, improve the curing and physical properties and sometime cheapen the final cost of rubber products [1, 2]. Considering the commercially available fillers, there are not many types of fillers that have been largely focused. Carbon black and silica are the example and are the representative for black and white fillers. These two fillers are frequently applied when the strength is required. However, to some extent, non-reinforcing fillers are recommended when other concerns such as the cost of the product and processing accounted. These include calcium carbonate, talc and clay, as well as other fillers from renewable resources and waste materials [3-5].

Nowadays, there has been a critical discussion about natural-based engineering products and this has been spreading interest worldwide. This is clearly responsible to latest issue concerning renewable resources. The context of sustainable development has been involved throughout the waste disposal strategy which is aimed to convert worthless materials into sustainable solution. In preparing rubber products, this term is also implemented. Many studies have focused and conducted on the use of natural fillers in the rubber compounds by both academicians and industrial researchers. One of the waste materials that is abundant is from lignocellulose materials. Studies on the utilization of this material in rubber matrix have been largely explored. As for example, Fiorote *et al.* [6] tried to modify the cellulose from empty fruit bunch and further used as filler for natural rubber, it was observed that the nanostructured particles of such material was successfully prepared and has greatly influenced the mechanical properties of natural rubber. Cellulose from kenaf fiber was also prepared by Roy and Potiyaraj [7], it was then compounded with natural rubber to observe the mechanical and dynamic properties of the composites. It was clear that the prepared cellulose had a direct effect in enhancing the properties of the composites.

Tea dust or tea waste (TW) is becoming a waste material especially in urban areas due to the increase in the consumption of tea globally. It was reported to reach about five million tons per year [6]. This has brought parallelly to the abundant TW. The TW is considered as a solid biomass waste and generally dispose into an environment. It is not being used in any purpose unless discarded as wet garbage. From this point of view, TW can be advantageous waste, if they are used in the application of rubber materials because they are priceless, light in weight, rich in polyphenols due to the tannin composed in tea leaves [8-10]. Although there are some attempts to utilize it in other industries such as textile industry, agricultural applications and in a production of animal feed, the effectively usage

of the TW is limited and the utilization of TW remains largely unexplored. Besides, TW also consists a high concentration of lignocellulose, this was evidently reported by Bernal *et al.* [11]. In this respect, the use of the TW as filler for natural rubber can be potential in a way to replace lignocellulosic filler.

The aim of this present study was to utilize TW as alternative filler for rubber compound. The study was evaluated through the curing characteristics, tensile properties and morphology of the composites. The results obtained from this study will bring an understanding on the role of TW as filler for natural rubber compound and extend to be source of crucial detail for preparing the rubber products based on TW filled natural rubber composites. To date, there is no single attempt has been made to investigate on the effects of TW contents on the properties of natural rubber composites. The solution presented in this work relates to the key benefits on producing new materials with economic, environmental and property-related aspects from TW mixed with natural rubber.

## 2.0 METHODOLOGY

### 2.1 Materials

Table 1 lists the compounding ingredients used to prepare the composites. The NR used was SMR 20 grade and was supplied by Mardec Malaysia Sdn. Bhd. (Selangor, Malaysia). The TW powder was supplied by Gold Leaf Marketing Sdn. Bhd. (Penang, Malaysia). Other compounding ingredients such as zinc oxide, stearic acid, N-cyclohexyl-2-benzothiazole-sulphenamide (CBS), 2,2 methylene-bis-(4-methyl-6-tert-butylphenol) (Vulkanox BKF) and sulfur were purchased from Bayer (M) Ltd.

**Table 1** Compounding ingredients for preparing the TW filled NR composites

Ingredients	Amount (phr)
SMR 20	100
Zinc Oxide	1.5
Stearic Acid	1.5
CBS	1.9
Vulkanox BKF	2.0
Sulfur	1.6
TW Powder	0 – 30

### 2.2 Preparation of TW Powder

TW is unable to be used as received, the size was reduced by grinding it using a Pulverizing Machine assembled from Rong Tsong Precision Technology Co. Ltd. (Dali District, Taichung City, Taiwan). Then, the obtained powder was sieved with an Endicott's sieve to get particle size below 100  $\mu\text{m}$ . Finally, TW was washed and dried in a vacuum oven at 105  $^{\circ}\text{C}$  for approximately 2 h to expel the moisture. The

resultant TW was now ready to be used as filler for NR. The morphology of TW was also captured and is shown in Figure 1. It can be clearly seen that TW is in irregular shape, varying in size and length. The particle size of TW is less than 250  $\mu\text{m}$  as observed in the SEM image.

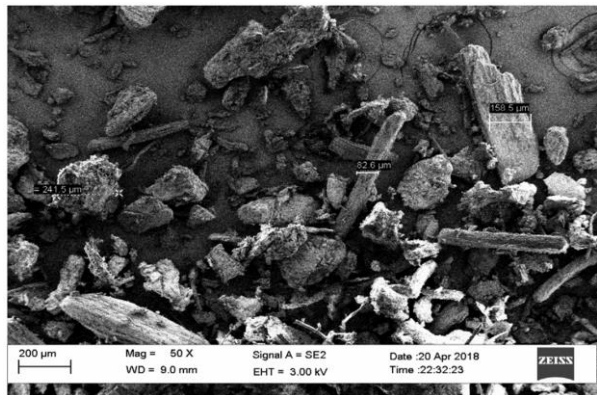


Figure 1 SEM image of TW powder at magnification of 50 $\times$

### 2.3 Compounding and Vulcanizing Steps

The compounding of NR, TW and other additives were done on a laboratory-sized (160  $\times$  320 mm<sup>2</sup> model XK-160) two-roll mill (Shanghai Rubber Machinery Works, Shanghai, China) at ambient temperature for approximately 20 min. The resulting compounds were then tested for their curing characteristics using a Monsanto Moving Die Rheometer (MDR 2000) (Alpha Technologies, Hudson, OH, USA) according to ASTM D5289. The test was carried out at 150°C and the outputs in term of scorch time ( $t_{s2}$ ), curing time ( $t_{c90}$ ) and torques were recorded automatically. Subsequently, the compounds were placed on the stainless steel mould and compressed at 150°C with the pressure of 10 MPa based on the respective curing times.

### 2.4 Fourier Transform Infra-Red (FTIR) Spectroscopy Analysis

The functionalities of TW were detected by using Fourier transform infrared spectroscopy (Perkin Elmer System 2000; PerkinElmer Inc., Waltham, MA, USA). The FTIR spectrum was obtained in the wavenumber range of 4000 to 550  $\text{cm}^{-1}$ . The TW was ground with potassium bromide (KBr) for the formation of pellet that has been used to obtain the infrared spectrum.

### 2.5 Measurement of tensile properties

The rubber vulcanizate sheet was first cut into dumbbell shape by using Wallace die cutter before undergoing the tensile testing. The tensile test was carried out by using Instron machine model 3366 (Instron Corporation, Norwood, MA, USA) according to ASTM D412 with a crosshead speed of 500 mm/min. The data obtained

from this test were the tensile strength, elongation at break and the modulus at specific strain or so-called tensile modulus.

### 2.6 Rubber-filler Interaction

A vulcanized rubber was cut into rectangular shape of 30 mm  $\times$  5 mm  $\times$  2 mm for every formulation. The sample was then soaked in toluene and kept swollen in a closed bottle for 72 h at room temperature. Afterwards, the sample was taken out and wiped quickly and weighed. Finally, it was dried and weighed again. Calculation of rubber-filler interaction ( $Q_f/Q_g$ ) was based on the Lorenz and Park's equations which is seen below [12].

$$\text{Rubber-filler interaction} = \frac{Q_f}{Q_g} \quad (1)$$

Where the  $f$  and  $g$  subscripts are referred to the filled and gum vulcanizates. While  $Q$  is the weight of toluene uptake per gram of rubber hydrocarbon was calculated by using the equation (2).

$$Q = \frac{W_2 - W_3}{W_1 \times \frac{100}{\sum phr}} \quad (2)$$

Where  $W_1$  is the original weight,  $W_2$  is the swollen weight and  $W_3$  is the dried weight. Low  $Q_f/Q_g$  value exhibits high rubber-filler interaction

### 2.7 Scanning Electron Microscopy

The morphological properties of the sample of tensile fractured surface and TW was studied by using Scanning Electron Microscopy (SEM) (Zeiss Supra-35VP; Carl Zeiss AG, Oberkochen, Germany). The sample from the broken tensile testing were collected and cut. The vulcanizates rubber sample and the TW then undergo sputter-coated process with a thin layer of gold to avoid electrostatic changing and poor image resolution.

## 3.0 RESULTS AND DISCUSSION

### 3.1 Fourier Transform Infra-Red (FTIR) Spectroscopy Analysis

Figure 2. There are some interesting absorbance peaks observed especially at wavenumbers of 3298, 2993, 1620 and 1032  $\text{cm}^{-1}$ . A broad band at a wavenumber of 3298  $\text{cm}^{-1}$  corresponds to O-H stretching obtained from the existence of alcohols and phenols in the TW. The peak at 2993  $\text{cm}^{-1}$  (medium) is due to the C-H stretch of TW. Another interesting peak is the amine group in the form of N-H bend and C-N stretch (aliphatic amines) available which are observed at the wavenumber of 1620  $\text{cm}^{-1}$

and  $1032\text{ cm}^{-1}$  respectively. The peaks observed are very good agreement to the previous work reported by Uddin *et al.* [8] and Tian *et al.* [13].

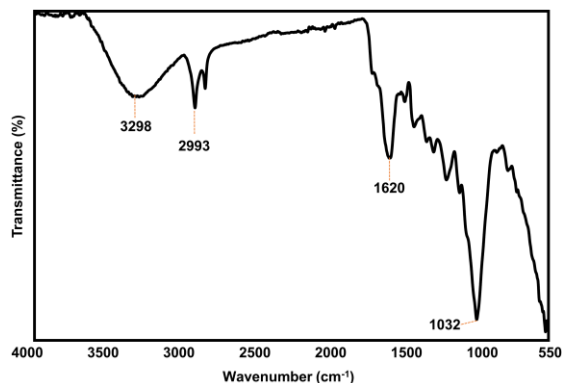


Figure 2 FTIR spectrum of TW powder

### 3.2 Curing Characteristics

Table 2 lists the curing characteristics of the TW filled natural rubber composites. It was observed that the maximum torque ( $M_H$ ) increased gradually with increasing the TW contents. The  $M_H$  value is defined as an elastic modulus and/or the hardness of the fully cured rubber [14]. A gradual increment of  $M_H$  value is simply responsible to a harder characteristic of the natural rubber composites as TW is in rigid form. This has brought to a decrease in the movement of the rubber chain and the flexibility of the chain thus allowing the increasing of the  $M_H$  value. The values observed are considered to be lower as compared to other types of fillers (e.g., carbon black, silica, clay and so forth). Because the nature of TW is not as hard as those fillers. Scorch time ( $ts_2$ ) showed a decreasing trend over the TW contents.  $ts_2$  is a time till the torque unit increases above the minimum which measures scorch safety time. Another definition of scorch time is also known as induction time and time where the crosslinks begins in the rubber compound. As can be seen from the FTIR spectrum of TW, it was found that there is an interesting peak of amine group available in TW. This has brought to an increase in pH of rubber compounds and in most instances, enhances the cure rate. Any chemical substance that gives the rubber compound more alkalinity will lead to enhance the cure rate because the acidic materials tend to retard the reactivity of accelerators [15, 16]. Therefore, it is expected that the amine-content in TW can accelerate the cure rate of the composites. A similar observation was also observed for the curing time ( $tc_{90}$ ) of the composites. The definition of cure time ( $tc_{90}$ ) is the time that required for the compound to reach 90% of the total cure during curing characteristics test. As expected, the available amine residing on the TW functionalities plays very important role in fastening the curing process of the natural rubber compound.

Table 2 Curing properties obtained from MDR 2000 of TW filled NR composites

TW contents (phr)	$M_H$ (dNm)	$ts_2$ (min)	$tc_{90}$ (min)
0	5.21	3.25	5.37
5	5.27	3.10	4.35
10	5.33	2.60	3.58
15	5.56	2.49	3.56
30	5.68	2.44	3.50

### 3.3 Tensile Properties

Tensile properties of TW filled natural rubber composites were also observed in this experiment. This is to find the optimum amount that is suitable to be used for natural rubber. Figure 3 shows the tensile strength and elongation at break of the respective composites. The tensile strength of the natural rubber composites decreased as the TW loading increased. This could be due to the incompatibility or poor adhesion between TW and matrix of rubber. On the other words, with increasing of TW loading, the possibility of TW to agglomerate increases therefore reduced the adhesion between rubber and TW and decreased the tensile strength. According to Xie *et al.* [17] and Ismail *et al.* [18], the hydrophilic filler in non-polar rubber can lead to the agglomeration of filler and later distribute unevenly throughout the matrix. Besides, insufficient wetting of TW throughout the matrix has also led to a weak interfacial adhesion, this later provides less efficiency of the stress to be transferred from the matrix to filler. Referring to Medalia [19] and Wang *et al.* [20], the filler aggregates are inadequate to be separate or adhered by the phase of rubber when the filler loading increased. Another possible reason is definitely due to the agglomeration of TW, this agglomeration enables to form a domain that acts like a foreign body and cause a non-homogenize dispersion.

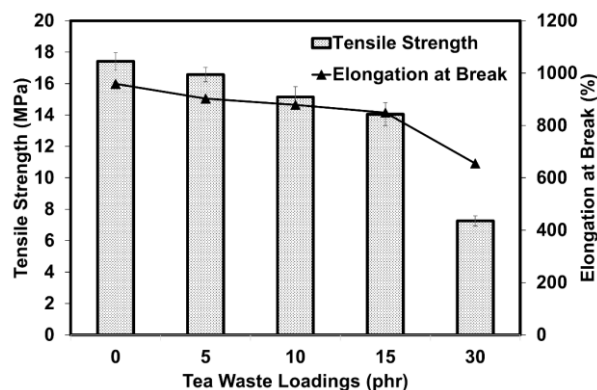
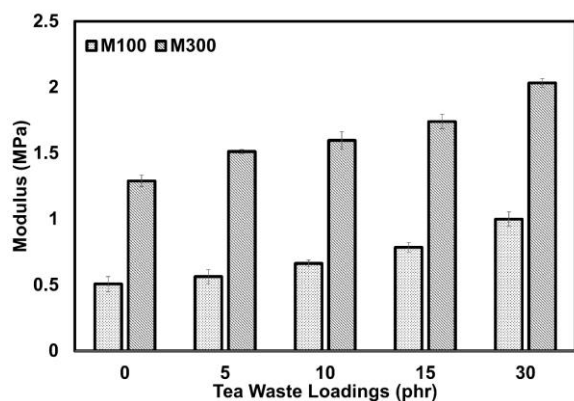


Figure 3 Tensile strength and elongation at break of TW filled NR composites



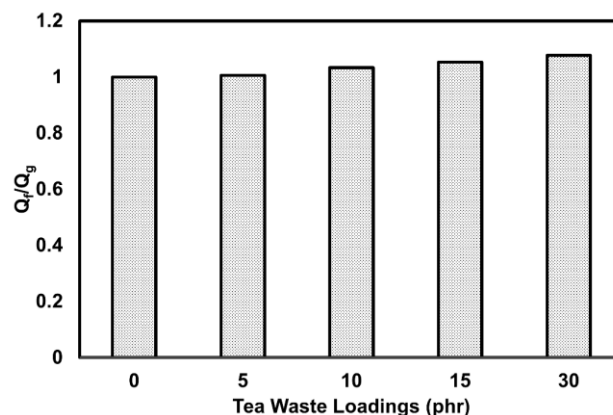


**Figure 4** Modulus at 100% (M100) and 300% (M300) strains of TW filled NR composites

Similar finding was also observed for the elongation at break of the composites. This could be due to the incorporation of unstrained particle of TW that leads to the formation of aggregates in between the chains of flexible rubber and restricts the mobility. The addition of filler may reduce the elasticity of the composites; higher TW loading has caused a stiffer natural rubber composites. As for the tensile modulus (see Figure 4), the stresses at 100% and 300% strains significantly increased with the addition of TW. When more TW gets into the natural rubber matrix, the composites become stiffer, the macromolecular chains of the rubber has the difficulty to move because of the restriction to the deformation hence increased the tensile modulus.

### 3.4 Rubber-filler Interaction

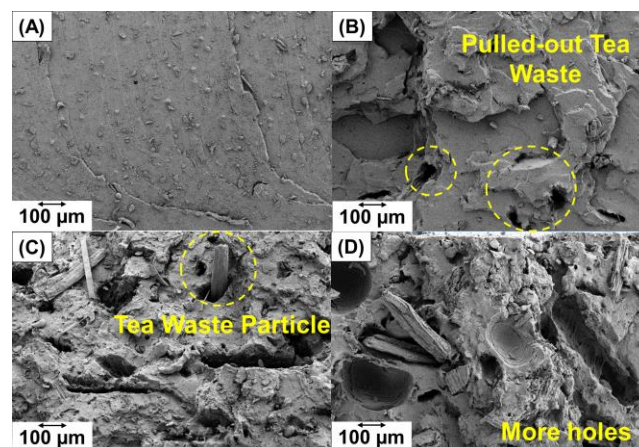
As can be seen in the preceding session, inclusion of TW to the natural rubber matrix has influenced the tensile strength of the composites due to its poor distribution of TW throughout the rubber matrix. To confirm such observation, the rubber-filler interaction of TW filled natural rubber composites was also carried out and is shown in Figure 5. The value of  $Q_f/Q_g$  slightly increased with increasing the TW contents. As widely known that that the higher value of  $Q_f/Q_g$ , the extent of interaction between the rubber and filler is lower. It can be concluded that the natural rubber composites that contains higher loading of TW have the weaker interaction between rubber and filler. Through this result, it is in a good agreement with the tensile strength observed in the previous study. Correlation between  $Q_f/Q_g$  values and the respective tensile properties has previously discussed elsewhere [21, 22].



**Figure 5**  $Q_f/Q_g$  values of TW filled NR composites

### 3.5 Morphological Characteristics of the Composites

The breaking samples from the tensile test were collected and screened for their morphology. The obtained images are shown in Figures 6A – 6D. Considering the unfilled vulcanizate (see Figure 6A), the morphology was more towards a flat and smooth surface due to the absence of filler in the matrix. However, when more TW get into the matrix, i.e., 5 – 30 phr (see Figures 6B – 6D), it revealed that increasing of TW contents, the fracture of composites become rougher. It is also seen that there is an evidence of filler agglomeration and uneven distribution of TW in the matrix of rubber. Apart from that, the images show a lot of TW pullouts from the matrix of rubber and some cavities are noticed. This is because of the poor adhesion between irregular shaped of TW particles and the matrix of rubber that cause the TW to be pulled out from the rubber easily when stress is applied hence leaving the gaping holes. This correlates to the lower tensile strength observed according to the previous results.



**Figure 6** SEM images at the magnification of 100× of TW filled NR composites i.e., 0 phr (A), 5 phr (B), 10 phr (C) and 15 phr (D)

## 4.0 CONCLUSION

This work aimed to study the potential use of TW as an alternative filler for natural rubber. The particle size of the TW used is less than 250  $\mu\text{m}$  as shown by SEM. It can be considered to be in a class of non-reinforcing filler. Based on the functionality observation of the TW, it consists of alcohols and phenols, carboxylic acid, amine and aromatic. Due to the presence of amine group available in TW, it can fasten the curing process of the composites, as the  $t_{s2}$  and  $t_{c90}$  reduced with increasing the TW contents. Adding the TW has increased the tensile modulus and  $M_H$  of the composites due to its stiffer and harder characteristics of the TW within the rubber matrix. However, the tensile strength and elongation at break of the composites drop continuously with the incorporation of the TW. This is simply due to several factors, i.e., the bigger size, incompatibility of filler and rubber and the distribution of TW throughout the matrix. Such findings can be verified by the  $Q_f/Q_g$  values, indicating less rubber-filler interaction when a higher amount of TW was used. Apart from that, SEM images also provided some evidence relating to the tensile properties observed. In summary, the TW at the loadings of 10 phr is highly suggested to be used as an alternative filler for natural rubber due to the overall satisfactory properties.

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