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# Water absorption and tensile properties of ground tire rubber/epoxy composites: effect of surface treatment

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**ABSTRACT**

This paper investigates the surface treatment effect of ground tire rubber (GTR) on the water absorption and tensile properties of aged GTR/epoxy composites. The GTR was treated using 10 wt-% NaOH solution and 1 wt-% stearic acid (SA) in acetone. The results showed that the GTR addition increased the water absorption and decreased the tensile strength and elastic modulus of epoxy composites. In dry condition, compared to the untreated GTR, the tensile strength of the treated GTR decreased by about 4.8% and 2.9% for the NaOH- and SA-treated GTR, respectively. Nevertheless, the addition of treated GTR increased the elastic modulus of epoxy composites by about 8.4% and 6.0% for the NaOH-and SA-treated GTR, respectively. For the aged epoxy, the tensile strength and elastic modulus decreased further due to the matrix plasticisation and degradation of the interface GTR/epoxy strength. It was found that the SA-treated GTR has better performance than the NaOH-treated GTR.

## Introduction

With increasing automobile production, the tire rubber demand considerably increases and this will lead to the generation of tire rubber waste. It was reported that in 2017 the world produced end-life tires (ETL) of about 29.1 million tons. Indonesia itself generated ETL of about 684.4 kilotons, and only 75% had been recovered for civil engineering, energy, and material recoveries [1]. Tire rubber has good properties and potential to be used as a filler of polymer, both thermoplastic and thermoset [2–8]. One of the thermoset polymers is epoxy, which has been extensively used in engineering structures. The successful incorporation of tire rubber waste into epoxy could reduce tire rubber waste and lower the material cost for developing high-performance epoxy.

Epoxy has good mechanical properties, chemical resistance, high bond strength, and low shrinkage, so it is widely used in many applications, including adhesives, coating, and matrix of advanced composite materials [9]. Introducing the second phase (hard and soft particles) into the epoxy can modify the physical and mechanical properties. Hard and soft particles can be used as filler. Hard particles can improve the stiffness of epoxy, but in contrast, soft particles, such as rubber, reduce epoxy stiffness. Rubber particles are mainly used for toughening agents of epoxy as they promote toughening mechanisms such as shear banding, which increases the toughness remarkably. Carboxyl terminated butadiene acrylonitrile (CTBN) is liquid rubber that can increase epoxy toughness

by factors 2–3 [10–12]. However, CTBN is expensive and difficult to control phase separation (i.e. particle size) during curing. Another particle is a preformed rubber particle. Preformed rubber particles are easier to process and control the particle size, and it has better performance than the CTBN [13–15]. The preformed rubber particles can be obtained from ground tire rubber (GTR) waste. However, surface treatment is required to increase the adhesion between GTR particles and the epoxy matrix, maximising the GTR role. The interaction between GTR and epoxy is mostly dominated by physical interaction such as van der Waals forces, so removing a weak boundary layer from the GTR surface could ensure intimate contact between GTR and epoxy, leading to enhanced adhesion. Several treatments have been reported to improve the surface energy of GTR, facilitating better adhesion with a matrix. Segre et al. [16] used NaOH for the surface treatment of GTR. NaOH solution was able to solute the zinc stearate from the rubber surface and improve its surface energy. Colom et al. [17] used sulphuric (96%), nitric (60%), and perchloric acids (60%) to treat GTR particles. Both sulphuric and nitric acids were able to improve the stiffness of the GTR and improve the rigidity of high-density polyethylene (HDPE). The sulphuric acid could increase the porosity and surface roughness of GTR, which then enhances the adhesion with the polyethylene matrix [18]. Besides using acid and base solution, the utilisation of a hybrid of sulphuric acid and silane to further increase the adhesion of the GTR/rubber

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matrix has been reported by Klajn et al. [19]. The increased adhesion between GTR and epoxy can improve the mechanical properties; however, in its application, the GTR/epoxy composites are often exposed in a moist environment. The absorbed water/moisture can affect the integrity of the GTR/epoxy interface, which then influences the performance of GTR/epoxy composites. Therefore, the effect of the absorbed water on the performance of GTR/epoxy composites is of research interest.

Water can ingress into the epoxy and then degrades the physical and mechanical properties via plasticisation, crack, and chemical degradation [20, 21]. The addition of fillers (hard and soft/rubber particles) affects the water absorption behaviour of epoxy. The water absorption can increase or decrease depending on the type of filler and content [22–24]. For inert fillers, the addition of fillers decreased the water uptake, but for reactive fillers, they increased the water uptake [23]. Hydrophobic fillers such as calcium carbonate, carbon black and graphene increased the water uptake but decreased the diffusion rate [25, 26]; therefore, proper surface treatment of filler is one of the key points to obtaining high water absorption resistance and mechanical properties of the filled polymer.

As mentioned in the previous paragraph, using a high concentration of acid and base solution for GTR surface treatment are effective but not environmentally friendly; therefore, low-cost and environmentally friendly GTR surface treatment is preferable. Moreover, the effects of GTR surface treatment on the tensile properties of GTR/polymer composites exposed in a moist environment are rarely reported. In this paper, the surface treatment of GTR using low alkali (NaOH) solution and stearic acid (SA) and their effects on the water absorption and tensile properties of GTR/epoxy composites in dry condition and after being aged in distilled water (wet condition) had been investigated. This study will benefit from utilising tire waste to modify epoxy used in polymeric matrix composites and adhesives, even if exposed to a moist environment.

## Materials and methods

### Materials

The epoxy resin and hardener were respectively a diglycidyl ether of bisphenol-A and a polyamidoamine, supplied by Justus Kimia Raya, Surabaya, Indonesia. The epoxy resin has an equivalent epoxy weight of $189 \pm 5$ g/eq, a viscosity of 13,000 MPa.s, and a density of 1.15 g/cm$^3$. The polyamidoamine has an amine hydrogen equivalent weight of 120 g/eq, a viscosity of 12,000 MPa.s, and a density of 0.97 g/cm$^3$ [25]. The GTR was made by scraping the tire rubber, screening and washing, and drying it to obtain a particle size of about 200 mesh (0.074 mm) or less. The composition of GTR (by wt-%) was rubber polymer (51%), carbon black (25%), oils (softeners) (19.5%), zinc oxide (1.5%), sulphur (1%), accelerators (0.5%), antioxidants, etc. (1.5%) [27].

### GTR surface treatment

The GTR was surface-treated using two methods: alkali (NaOH) and stearic acid (SA) treatments. For the NaOH surface treatment, the GTR was immersed in 10 wt-% alkali (NaOH) solution for 30 min. The concentration of NaOH was based on published papers [28, 29]. After the surface treatment, the GTR was rinsed and dried in an oven at a temperature of 60°C. For the SA surface treatment, the GTR was immersed in 1 wt-% SA in acetone. This concentration was adopted from Ref. [30]. After this treatment, the GTR was dried in the oven at a temperature of 60°C. The untreated (UT) GTR was used as a control.

### Fabrication of tensile specimens

To manufacture the tensile specimen, the GTR particles were mixed with epoxy resin with a content of 10 wt-%. The mixture was stirred using a high-speed mixer for about 10 min to obtain a homogeneous mixture. It was then followed by degassing in a vacuum chamber to remove the air bubble trapped during stirring. Hardener was added to the mixture and slowly stirred by hand for about 5 min, and then was degassed again for about 30 min. The epoxy resin to hardener ratio was 3:2 (by weight). The ratio was selected as it has low water absorption and high mechanical properties. The mixture was then cast into the dog bone-shaped silicon mould according to ASTM D638 [31] and left to cure for at least 24 h.

### Fourier transform infrared spectroscopy (FTIR)

The FTIR was carried out to investigate the surface chemistry of the untreated and treated GTR particles and GTR/epoxy composites in dry and wet conditions. The FTIR was conducted using a Perkin Elmer Frontier Fourier Transform Infrared Spectrometer. The weight of the sample used in the test was approximately 0.2 g in the KBr pellets. The spectra were recorded in the wavenumber range of 4000–400 cm$^{-1}$.

### Water absorption

Water absorption was carried out using the tensile specimen directly. The specimens were immersed in distilled water at a temperature of 50°C. Before the immersion, the specimens were dried in an oven at a temperature of 50°C for 24 h. This dried tensile specimen was weighed to obtain the initial weight of the

specimen before being immersed in distilled water. During immersion, the specimens were taken periodically for weighing. The weighing was performed using a Kenko digital microbalance. The weighing was conducted 3 times (after 3, 10, and 17 h since being immersed) for the first day and once for the following days up to an aging time of 75 days before the tensile testing in the wet condition. After the aging time of 75 days, the specimen was estimated to reach the saturation level.

### Tensile testing

The tensile test was carried out on the dry specimen (unaged) and the wet (aged) specimens. For the wet test, the specimen was aged until closing or reaching the equilibrium state, where the degradation due to water content was possibly maximum, for the worst case. Literature reported that the degradation of epoxy mechanical properties due to water was essentially linear [32, 33], so the water-dependent mechanical properties could be predicted by interpolation. The tensile test was conducted using a Tensilon universal testing machine (10 kN load capacity) with a displacement rate of 5 mm/min. An extensometer was used to measure the elongation. Three replications were made for each test.

### Scanning electron microscopy

After the tensile test, the fracture surfaces were examined using a scanning electron microscope (SEM) for morphology study. The SEM was carried out using a Hitachi Flexsem 100 SEM machine. A gold coating

was sputtered-coated on the fracture surface before being examined in the SEM machine.

## Results and discussion

### FTIR analysis

Figure 1 shows the FTIR spectra of the untreated and treated GTR particles. The peaks of transmittance are indicated in the figure. Generally, the transmittance spectra of the untreated and treated GTR spectra seem similar. The peak at a wavenumber of 3450 $cm^{-1}$ is associated with O–H stretching. If compared to the untreated GTR, the transmittance of the NaOH-treated GTR at that peak decreased (the absorbance increased), which may be due to the additional bond of –OH. This –OH bond increased the hydrophilicity of the NaOH-treated GTR. Meanwhile, for the SA-treated GTR, the transmittance at that peak tended to fade, indicating less -OH on the GTR surface. The peaks at 1633 and 1455 $cm^{-1}$ are associated with C = C stretching and C–H bending, respectively [34, 35]. The peaks at wavenumbers 1455, 1298, 1124 and 1010 $cm^{-1}$ are all associated with the S = O stretching, while the peak at 712 $cm^{-1}$ is associated with C–S stretching [34, 35]. The peak at 2350 $cm^{-1}$ is associated with carbon dioxide ($CO_2$) from the background. For the NaOH-treated GTR, the peaks at 1298 and 1124 $cm^{-1}$ are diminished or faded, indicating that the NaOH might have oxidised or opened the double bonds or broken the cross-linking [16]. Meanwhile, the stearic acid in acetone might have cleaned the GTR surfaces from contaminants and added a layer of carbonyl functional group after drying, indicated by the increased peaks at 1703 and 1535 $cm^{-1}$ [36]. In
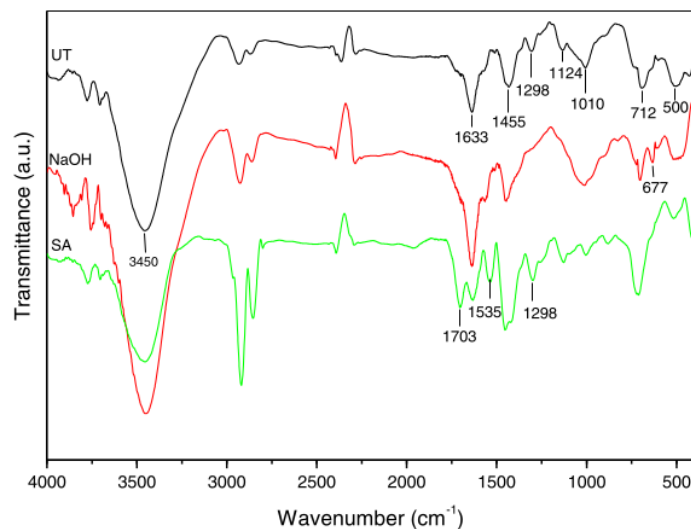


**Figure 1.** FTIR spectra of untreated and treated GTR particles.

this case, the stearic acid-treated GTR became hydrophobic. This was supported by a simple water drop test on the compacted GTR to measure the contact angle. The results indicated that the contact angle of SA-treated GTR and the untreated GTR was about 124.6° and 93°, respectively (see Figure 2).

Figure 3 shows the transmittance spectra of the GTR/epoxy composites in dry and wet (having aged in distilled water at 50°C for 75 days) conditions. The FTIR spectrum of neat epoxy is shown for comparison. The peaks of the FTIR spectra of composites were weaker than that of neat epoxy. This is related to the concentration of the samples. The transmittance decreases (the absorbance increases) when the sample concentration increases [37]. The FTIR spectra in the composites were dominated by the matrix, so the weaker spectra in composites could be due to the lower epoxy content in composites (90 wt-%). In general, the FTIR spectra of GTR/epoxy composites were similar to that of the neat epoxy, both in dry and wet conditions. The peaks at the wavenumber of 3450, 1630, 1510, 1240, 1113, 850–550 $cm^{-1}$ were associated

with the O–H stretching, C=C stretching, N–O stretching, C–N stretching, C–O stretching, C=C bending, C–H bending, and C-halogen stretching, respectively [35]. There were no disappearing or additional peaks observed in the spectra with the addition of GTR, indicating no chemical bonding between GTR and epoxy. In the composites, the signal of 2350 $cm^{-1}$ was reversed compared to those of neat GTR and neat epoxy, which might be due to the $CO_2$ content during infrared radiation [38]. If the $CO_2$ content increase after background measurement, the peak of transmittance decreases (absorbance decrease) and otherwise, if the $CO_2$ content increases after the background measurement, the peak of transmittance decreases (absorbance increases). Nevertheless, that peak is not of interest in this study.

In the wet condition, the attention should be focused on the peak at 3450 $cm^{-1}$, as it is related to hydroxyl (–OH) groups from the absorbed water in the composites. It is seen that in the wet condition, the transmittance peaks at 3450 $cm^{-1}$ for the untreated and NaOH-treated GTR/epoxy composites
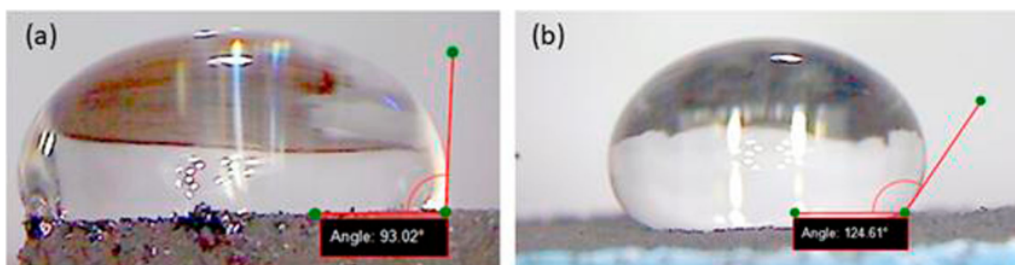


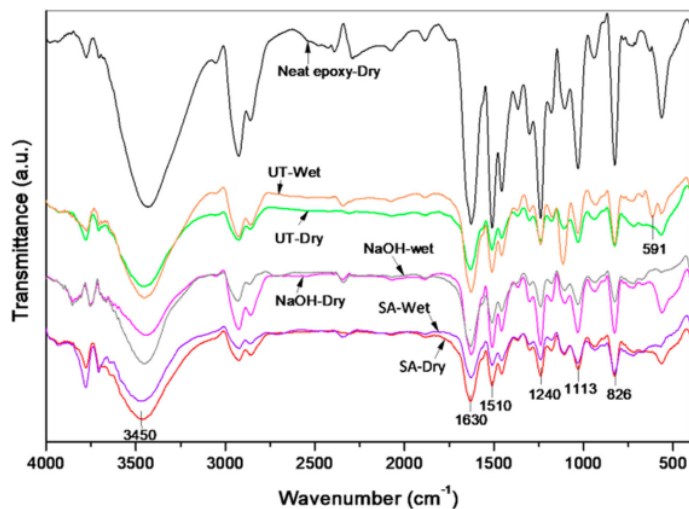**Figure 2.** Contact angle of (a) untreated and (b) SA-treatedGTR.



**Figure 3.** FTIR spectra of untreated and treated GTR/epoxy composites in dry and wet conditions.

decreased, indicating much absorbance of the infrared radiation. This indicated higher water content in the GTR/epoxy composites. However, the anomaly occurred on the SA-treated GTR/epoxy composites, where the peaks of the wet composites were weaker than that of the dry composites. The water repellence of hydrophobic GTR may take a role in this anomaly; however, it requires further investigation.

### Water absorption

Figure 4 shows the water uptake of GTR/epoxy composites vs the thickness-normalised square root of time at the different GTR surface treatments. The water uptake of neat epoxy is also shown for comparison purposes. It can be seen that the water uptakes of all GTR/epoxy composites and neat epoxy follow the Fickian diffusion behaviour. Initially, water uptake ($M_t$) increased proportionally with the square root of time ($\sqrt{t}$) and then slowed down, closing to the equilibrium water uptake ($M_{\infty}$). The diffusion rate ($D$) was calculated at the linear portion of the water uptake vs the square root of the time curve using Equation (1)

$$D = \frac{\pi}{16}\left(\frac{m \times l}{M_{\infty}}\right)^2 \qquad (1)$$

where $m$ is the slope of the linear portion of water uptake vs the square root of time data and $l$ is the sample thickness. The Fickian's water uptake was fitted against the experimental water uptake data following Equation (2) [39],

$$\frac{M_t}{M_{\infty}} = 1$$
$$-\frac{8}{\pi^2}\sum_{n=0}^{\infty}\frac{1}{(2n+1)^2}\exp\left[\frac{-(2n+1)^2\pi^2 Dt}{l^2}\right]$$
$$(2)$$

As seen in Table 1, the equilibrium water uptake of NaOH-treated GTR/epoxy composites was the highest, while the equilibrium water uptake of untreated and
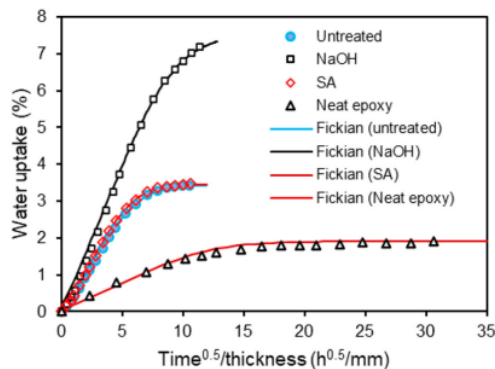
SA-treated GTR/epoxy composites was about the same and much lower than that of NaOH-treated GTR. However, water uptakes of all the GTR-filled epoxies were higher than that of the neat epoxy, about 80, 294, and 82% for the untreated, NaOH-, and the SA-treated GTR, respectively. This indicated that the existence of GTR considerably increased the water uptake of epoxy. It is worth noting that the equilibrium water uptake of NaOH-treated GTR/epoxy composites had not been reached when the weighing was stopped. However, using the Fickian diffusion model, the predicted equilibrium water uptake shown in Table 1 was in good agreement. The increase in water uptake of epoxy due to the addition of GTR could be explained by free volume and the filler/matrix affinity [40]. When the free volume and filler/matrix affinity increase, the water uptake tends to increase [40]. So, the increase in water uptake of the GTR/epoxy composites was likely due to the rise of free volume. The addition of GTR could reduce the molecular packing of the chain structure of epoxy, increasing free volume. Besides, the aggregation of GTR particles in the epoxy created voids that increased the free volume, as shown in Figure 5. In this case, because the surface characteristics of GTR after the different surface treatment was different, so the GTR/epoxy affinity and the dispersion of GTR were also different. It seemed that the NaOH treatment induced the GTR to be hydrophilic and had low dispersion, as shown by the GTR agglomeration (Figure 5b, marked by the dashed red circles). Meanwhile, the dispersion of the untreated and SA-treated GTR in epoxy was better than that of NaOH-treated GTR (see Figure 5a and c, respectively). The agglomerated GTR particles created voids and increased free volume, which became sites for water clustering. Therefore, it absorbed significant water compared to the untreated and SA-treated GTR. During aging, the surface of NaOH-treated GTR/epoxy composites blistered, considerably increasing the water uptake. The similar water uptake behaviour of the untreated and SA-treated GTR/epoxy composites indicated that the surface chemistry of GTR was quite similar, as shown in the FTIR spectra (Figures 1 and 3). To further clarify this behaviour, a water drop test was conducted on the GTR/epoxy composites. The contact angle was measured, and the results are shown in Figure 6. The contact angle of the NaOH-treated GTR/epoxy composite was lower



**Figure 4.** Water uptake of treated GTR/epoxy composites.

**Table 1.** Water absorption properties of GTR/epoxy composites.

| Specimen | $M_{\infty}$ (%) | $D \times 10^{-6}$ (mm²/s) |
|---|---|---|
| Neat epoxy | 1.90 | 0.36 |
| Untreated GTR/epoxy composites | 3.42 | 1.30 |
| NaOH-treated GTR/epoxy composites | 7.50* | 0.63* |
| SA-treated GTR/epoxy composites | 3.46 | 1.45 |

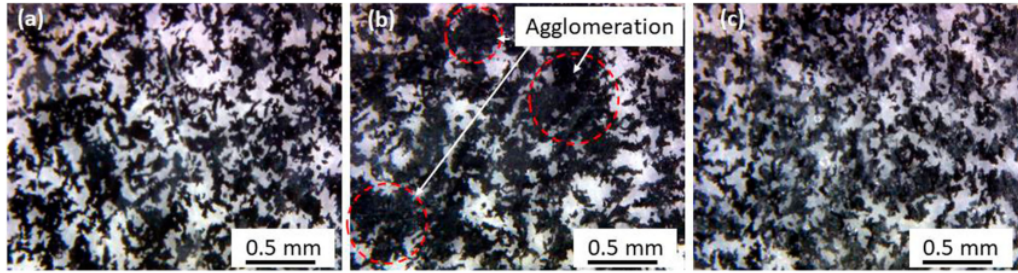Note: * predicted values using the Fickian diffusion model.

**Figure 5.** Distribution of (a) untreated, (b) NaOH-, and (c) SA-treated GTR in epoxy. All images were taken using a light microscope.

than those of the untreated and SA-treated GTR, while the contact angle of the untreated and SA-treated was about the same. The results seem to correspond to the equilibrium water absorption of NaOH-treated GTR/epoxy composites, which was the highest, while the equilibrium water uptake for the untreated and SA-treated GTR/epoxy composites was about the same. Thus the contact angle could be a good indicator of the water absorption behaviour.

The diffusion rate, as seen in Table 1, also increased with the addition of GTR into the epoxy. The diffusion rate is affected by the tortuosity of the diffusion path in the epoxy and the number of functional groups that attract water molecules during diffusion. Normally, the diffusion rate is less for the hydrophilic filler introduced in the epoxy [41]. The neat epoxy had the lowest diffusion rate among the epoxy systems, followed by the NaOH-, untreated, and SA-treated GTR/epoxy composites. The low water uptake and the diffusion rate of the neat epoxy were likely due to its high cross-link density and less free volume. The addition of GTR could reduce the cross-link density and increase free volume in epoxy, indicated by the increased water uptake and diffusion rate. The lower diffusion rate of NaOH-treated GTR/epoxy composites compared to the untreated and SA-treated GTR was highly likely due to its hydrophilic nature, which has more functional groups.

**Tensile properties**

Figure 7 shows the typical stress–strain curves of GTR/epoxy composites. The stress–strain of neat epoxy

(NE) was shown for comparison. In the dry condition, the stress–strain curves were elastoplastic. The softening was observed for the neat epoxy and the untreated GTR/epoxy composites. However, the stress at break of the treated GTR occurred at the ultimate stress, and the strain at break of the treated GTR/epoxy composites was lower than those of the untreated GTR/epoxy composites and neat epoxy. In the wet condition, the stress–strain curves pattern of neat epoxy seemed similar to that in the dry condition; however, the stress–strain curves of the untreated and SA-treated GTR/epoxy composites tended to be elastic-perfectly plastic, except for the NaOH-treated GTR/epoxy composites. The plasticisation of the matrix contributed to the increase of strain at break for the
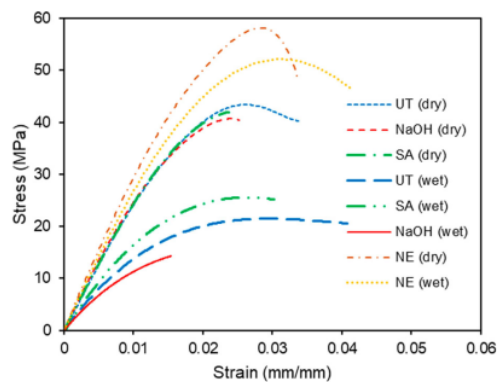


**Figure 7.** The typical stress–strain curves of GTR/epoxy composites in dry and wet conditions.
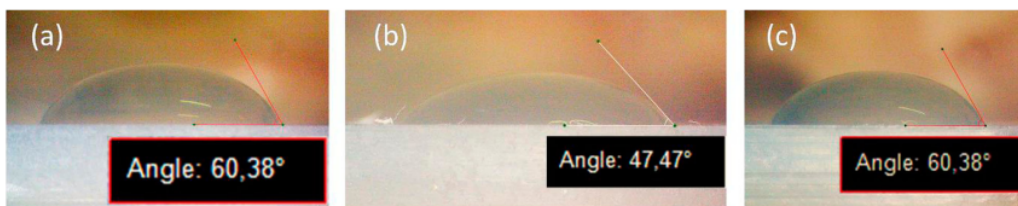


**Figure 6.** Contact angle of (a) untreated, (b) NaOH-, and (c) SA-treated GTR/epoxy composites.

aged specimen compared to the dry specimens. However, the low strain at the break of NaOH-treated GTR/epoxy composites was probably due to the voids formed in the GTR/epoxy interface during aging. The voids acted as stress raisers, initiating premature failure.

Figure 8 shows the tensile properties (the tensile strength, elastic modulus, and the strain at break) of GTR/epoxy composites and neat epoxy. In the dry condition, the tensile strength of GTR/epoxy composites was lower (about 25–28%) than that of the neat epoxy. It is expected as the GTR is soft and inferior to the epoxy tensile strength. The tensile strength (Figure 8a) of the treated rubber/epoxy composites was slightly lower than the untreated GTR, about 4.8% and 2.9% for NaOH- and stearic acid-treated GTR, respectively. If the tensile strength in dry and conditions was compared, the decrease of the tensile strength in wet conditions was about 11%, 48%, 64%, and 42% for neat epoxy, untreated, NaOH-, and SA-treated GTR/epoxy composites. In the wet condition, compared to the wet epoxy, the decrease

of the tensile strength was higher than those in the dry condition, about 55%, 71%, and 52% for the untreated, NaOH-, and SA-treated GTR/epoxy composites. In the wet condition, besides the plasticisation of the matrix, the weakening interface GTR/epoxy strength also contributes to the decrease of the tensile strength. In this case, the decrease in tensile strength of NaOH-treated GTR/epoxy composites was the highest. This is likely due to the degradation of the interface strength after absorbing much water. Water attacked the interface of GTR/epoxy and displaced the matrix from the GTR. It is seen that the SA-treated GTR gave a better improvement of the tensile strength of GTR/epoxy composites in the wet condition compared to the others. This could be attributed to the hydrophobic nature of SA-treated GTR, which improved the dispersion and bonding strength of the GTR/epoxy interface. Other researchers, Sipahi-Saglam et al. [42] reported the increased tensile strength of epoxy filled with GTR after treating the GTR using acrylic acid and acrylic acid/benzoyl peroxide mixture. The improved interfacial strength of
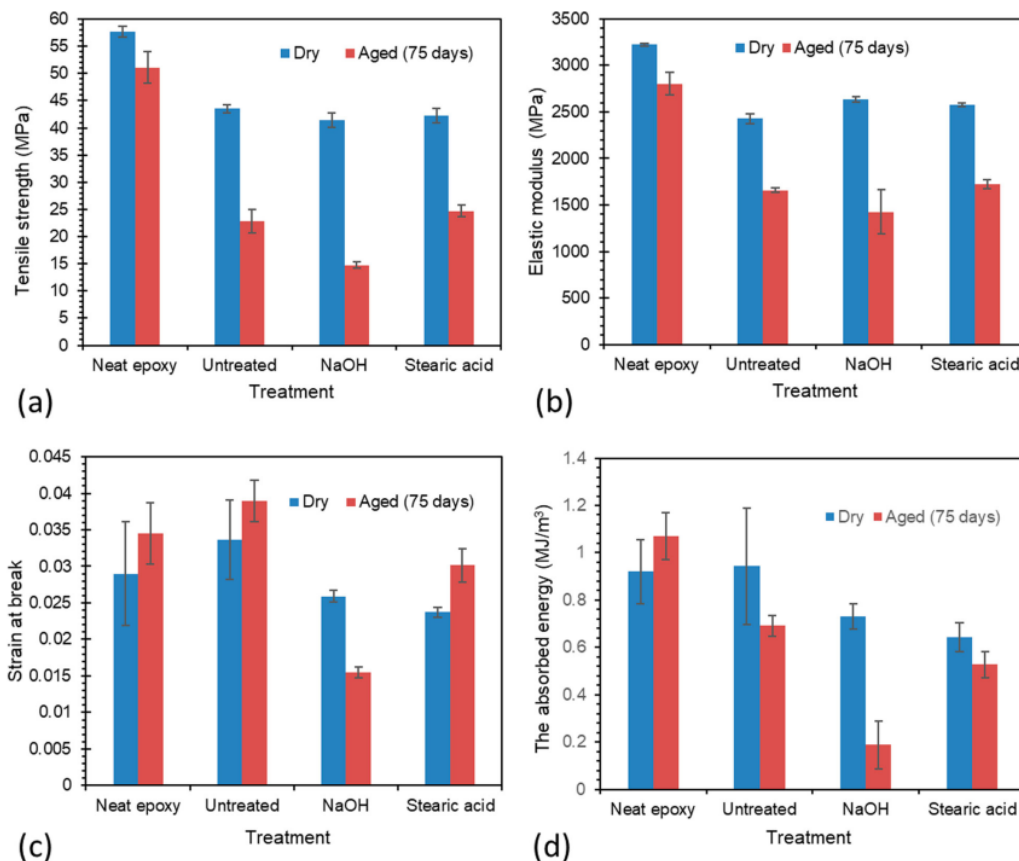


**Figure 8.** Showing the tensile strength (a), elastic modulus (b), the strain at break (c), and the absorbed energy (d) of GTR/epoxy composites in dry and wet conditions.

the surface-treated GTR/epoxy interface was also reported by Kaynak et al. [43]. However, they used vinyltriethoxysilane, 3-aminopropyltriethoxysilane, oxygen plasma with vinyltriethoxysilane, acrylic acid, and acrylic acid/benzoyl peroxide mixture to treat the GTR.

As seen in Figure 8(b), in the dry condition, the elastic modulus of the GTR/epoxy composites was lower, about 25%, 18%, and 20% for the untreated, NaOH-, and SA-treated GTR/epoxy composites, respectively, compared to the neat epoxy. Meanwhile, the elastic modulus for the treated GTR was higher than the untreated one, about 8.4% and 6.0% for the NaOH- and stearic acid-treated GTR, respectively. The NaOH treatment increased the GTR hardness [44], which might increase the stiffness of NaOH-treated GTR/epoxy composites more than the SA-treated GTR/epoxy composites. In the wet condition, the elastic modulus of the GTR/epoxy composites much further decreased compared to that of neat epoxy, about 41%, 49%, and 38% for the untreated, NaOH-, and SA-treated GTR/epoxy composites, respectively. Again, the SA-treated GTR reduced the degradation of elastic modulus better than the other treatments. It seems that the improved elastic modulus of SA-treated GTR/epoxy composites was due to the water resistance interface of the SA-treated GTR and epoxy. This is not the case for the NaOH-treated GTR, which is prone to water uptake attack, and its elastic modulus decreased more than the SA-treated GTR/epoxy composites.

Figure 8(c) shows the strain at the break of the GTR/epoxy composites and neat epoxy in dry and wet conditions. In the dry condition, the average strain at the break of neat epoxy was lower than that of the untreated GTR/epoxy composites, but it was higher than those of the treated GTR/epoxy composites. The strain at break of the treated GTR/epoxy composites was lower than the untreated one. It is consistent with the trend of the elastic modulus, showing that the treated GTR/epoxy composites became stiffer. Compared to the untreated GTR, the reduction of the strain at the break of the NaOH and stearic acid-treated GTR is 23.1% and 29.5%, respectively. In the wet condition, again, the average strain at the break of the untreated GTR/epoxy composites was higher than that of the neat epoxy. The addition of the treated GTR reduced the strain at break. However, as expected, the strain at break of all epoxy systems in the wet condition was higher than those in the dry condition, except that of the NaOH-treated GTR/epoxy composites. The increase of strain at break in the wet condition was attributed to plasticisation of the matrix, but the lower strain at break of the NaOH-treated GTR/epoxy composites might be due to the weak interface of GTR/epoxy and voids. Figure 8(d) indicates the absorbed energy for fracture (tensile toughness) of

the composites. The absorbed energy was calculated from the area underneath the stress–strain curves. In the dry condition, the trend of the absorbed energy for fracture was similar to that of the strain at break. The absorbed energy of the treated GTR/epoxy composites was lower than that of the untreated GTR as a consequence of the lower strain at break; meanwhile, the tensile strength was not much different. In the wet condition, although the strains at break of the untreated and SA-treated GTR/epoxy composites were higher than those in the dry condition, the absorbed energies were much lower than those in the dry condition. This was due to the low tensile strength in the wet condition. The very low absorbed energy of the NaOH-treated GTR/epoxy composites in the wet condition was due to the existence of voids at the surface (blistering) that decreased both the tensile strength and strain at break.

### Morphology of fracture surfaces

Figure 9 shows the SEM micrographs of tensile fracture surfaces of GTR/epoxy composites. In the dry condition, voids are observed for the untreated and the NaOH-treated GTR, indicating a weak interfacial strength (Figures 9a and b). In contrast, voids were not observed in the fracture surfaces of the SA-treated GTR particles (Figure 9c), indicating a good interfacial bond between the SA-treated GTR and the epoxy matrix. As blown up in Figure 9(c1), the SA-treated GTR might act as crack bridging, shown by the rough surface of the fracture GTR. These micrographs are consistent with the trend of elastic modulus; for example, the elastic modulus of the stearic acid-treated GTR/epoxy composite was higher than the untreated GTR/epoxy composites. It can be observed from Figure 6 that the GTR particles acted as crack deflectors, shown by rough fracture surfaces. In the wet condition, the SEM micrographs were shown in Figures 9 (d and e) for the untreated and NaOH-treated GTR/epoxy composites, respectively. The fracture surfaces of both seemed smoother than those in the dry condition, indicating the plasticisation of the epoxy matrix.

### Conclusion

The effects of GTR surface treatment on the water absorption and tensile properties of GTR/epoxy composites in dry and wet conditions had been undertaken. The surface treatment used 10 wt-% NaOH solution and 1 wt-% stearic acid in acetone. The addition of GTR into the epoxy increased the water absorption of the composites significantly compared to that of the neat epoxy. A remarkable increase in water absorption was shown for the NaOH-treated GTR due to its hydrophilic nature. In contrast, the water absorption
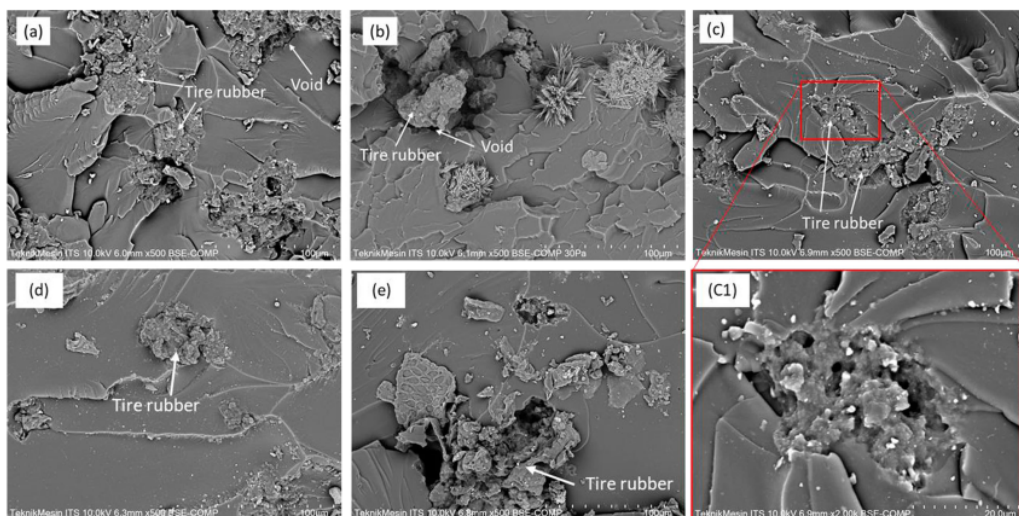
**Figure 9.** The SEM micrographs of the broken tensile specimens, for (a) untreated, (b) NaOH-, and (c) stearic acid-treated GTR/epoxy composites in the dry condition, while images (d) and (e) are respectively for the untreated and NaOH-treated GTR/epoxy composites in the wet condition.

behaviour of untreated and stearic acid-treated GTR was relatively similar and lower than that of the NaOH-treated GTR. Both in the dry and wet conditions, the addition of GTR into epoxy decreased their tensile strength and elastic modulus. The tensile strength of the treated GTR seemed slightly lower than that of the untreated GTR/epoxy composites. However, the elastic modulus of the treated GTR was higher than that of the untreated GTR. The strain at break of the treated GTR was lower than the untreated one, consistent with the elastic modulus increase. As expected, the strain at break in the wet condition was higher than in the dry condition except for the NaOH-treated GTR/epoxy composites. A better interfacial strength was observed for the stearic acid-treated rubber than for the NaOH-treated and untreated GTR. However, it requires further investigations on the fracture behaviour of GTR/epoxy composites.

## Disclosure statement

## Funding

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