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Reinforcement of Maleated Polyethylene/Ground Tire Rubber Thermoplastic Elastomers using Talc and Wood Flour

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ABSTRACT: Maleated polyethylene (MAPE)/Ground tire rubber (GTR) thermoplastic elastomer with 50 vol % GTR was reinforced by incorporation of talc powder and wood flour. Scanning electron microscopy (SEM) reveals that maleated polyethylene (MAPE) has good compatibility with wood flour, but the adhesion with talc particles is weak. Tensile moduli of MAPE/GTR increase more significantly after inclusion of talc particles compared to wood flour. Prediction of the tensile modulus of hybrid MAPE/GTR/particle composites is successfully performed using a combination of Kerner and Halpin-Tsai models. Elastic moduli are shown to depend strongly on both aspect ratio and level of particle dispersion in the matrix. Measurement of compression sets shows that elastic recovery of the compounds decreases after addition of solid particles. Samples having better particles/matrix compatibility show higher elastic recovery. Thermogravimetric analysis shows that inclusion of wood flour decreases thermal stability of compounds. Density and hardness of MAPE/GTR are also shown to increase after inclusion of particulate reinforcements. © 2013 Wiley Periodicals, Inc. J. Appl. Polym. Sci. **2014**, *131*, 40195.

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INTRODUCTION

Recycling of tire rubber has attracted a great deal of attention over the past decades due to environmental concerns. Vulcanized structure of tire rubber makes it impossible to melt or dissolve, which gives rise to very challenging recycling conditions. Fabrication of GTR from tire waste is proposed as the most promising method for reprocessing such materials.[1–7] Submillimeter fractions of GTR can be introduced into different polymeric matrices such as thermoplastics, thermosets, and even rubbers, to reduce their cost and modify their characteristics. Among these, thermoplastic/GTR compounds benefit from the elastic behavior of rubber combined with easy processing and reprocessing conditions of thermoplastics.[8–10]

Since rubber molecules in GTR are already crosslinked, they do not have the freedom to entangle with thermoplastic molecules. This behavior leads to a lack of compatibility between GTR and matrix phases causing poor mechanical properties of GTR based thermoplastic elastomers.[11–14] Several modification methods such as surface treatment and devulcanization have been proposed to increase the interactions at the interface of such compounds. Partial devulcanization (regeneration) of tire rubber has been performed via different techniques, while none was proven to be effective.[15] This is especially true for compounds having higher GTR concentrations. Previous investigations performed by the authors showed that MAPE, on the other hand, is an excellent choice for production of GTR-filled thermoplastic elastomers.[16] MAPE is assumed to create strong chemical bonds with vulcanized rubber through reaction of maleic anhydride groups with unsaturated (C=C) bonds of rubber molecules.[17–21] It was shown that compounds with GTR concentrations up to 90% by weight can be successfully produced using MAPE.[16]

Inclusion of a rubber phase, both virgin and recycled, to thermoplastics is known to result in noticeable reduction in mechanical properties, especially elastic modulus. This behavior is ascribed to low mechanical properties of rubbers in comparison with thermoplastics. In our previous work, it was reported that adding 70 wt % of GTR to MAPE decreased its tensile modulus by 88%.[16]

One effective method to improve the modulus of thermoplastic elastomers is addition of a reinforcing phase; i.e., particles or short/long fibers. Fiber reinforcement is known to increase the elastic modulus more significantly than particles, but processing of long fiber composites, on the other hand, is more challenging

Additional Supporting Information may be found in the online version of this article.

compared with particulate composites. Stiff particles, both organic and inorganic, have been frequently proposed in order to balance the mechanical properties of thermoplastics.[22–24] GTR and particulate reinforcements can easily be incorporated into thermoplastics for products having a wide range of applications, while mechanical properties can be controlled by altering filler concentration.

In this article, reinforcement of GTR-filled thermoplastic elastomers is performed through inclusion of rigid (organic and inorganic) particles with different origins, namely wood flour and talc. The morphological, mechanical, and physical properties of these ternary composites are then studied to evaluate the effect of each particle. Adding both particles is shown to increase elastic modulus of compounds, while talc was more effective. Prediction of the elastic modulus of MAPE/GTR compounds and hybrid MAPE/GTR/particle composites is also performed using Kerner and Halpin-Tsai models, respectively, with reasonable accuracy. Calculation of parameters of Halpin-Tsai model proved that elastic moduli of composites depend strongly on both aspect ratio and level of particle dispersion in the matrix.

THEORY

For composites containing both rigid particles and soft elastomeric particles, two limiting cases with different mechanical properties have been suggested by Jancar and Dibenedetto[25]:

i. perfect separation between the dispersed phases (rubber and rigid particles),
ii. perfect encapsulation of rigid particles by the rubber phase.

In our case, since GTR particles are vulcanized, no encapsulation of the rigid particles by the rubber phase can be expected. Thus, our MAPE/GTR/particle composites can be considered as a clear example of case (i). According to Jancar and Dibenedetto, in case (i), the elastic modulus of the thermoplastic/rubber blend (E_{TPE}) can be first predicted using Kerner's model.[25] In the case of perfect adhesion between the thermoplastic matrix and the "spherical" rubber particles, Kerner's model predicts the elastic modulus as:

$$\frac{E_{TPE}}{E_m} = \frac{(1+AB\Phi_{GTR})}{(1-B\Phi_{GTR})} \quad (1)$$

$$A = \frac{(8-10\upsilon_m)}{(7-5\upsilon_m)} \quad (2)$$

$$B = \frac{\left(\frac{E_{GTR}}{E_m}-1\right)}{\left(\frac{E_{GTR}}{E_m}+A\right)} \quad (3)$$

where E_m and υ_m are the elastic modulus (98.5 MPa) and Poisson ratio (0.5) of the MAPE matrix, respectively. E_{GTR} and Φ_{GTR} are the average elastic modulus, taken here as 2.0 MPa, and volume fraction of GTR particles, respectively.

For case (i), the elastic modulus of hybrid MAPE/GTR/particle composites can then be calculated as a two phase system including MAPE/GTR blend as the effective matrix and rigid particles as reinforcement. Here, the Halpin-Tsai equation is used to predict the elastic modulus of MAPE/GTR/particle composites which is one of the most frequently used models to predict the elastic modulus of composites as[26]:

$$\frac{E_c}{E_m} = \frac{1+\xi\eta\Phi_f}{1-\eta\Phi_f} \quad (4)$$

where

$$\eta = (m-1)/(m+\xi) \quad (5)$$

In eq. (5), m is the modulus ratio ($=E_p/E_m$), while E_p and E_m are the elastic modulus of particles and effective matrix ($=E_{TPE}$), respectively. Since both particles can be considered rigid compared with the matrix, E_p is much higher than E_m, therefore η tends towards unity ($\eta = 1$). ξ is a shape factor and depends on orientation and aspect ratio of the reinforcement. It is suggested that a good prediction for longitudinal modulus of composites (with perfect orientation of reinforcements) can be obtained using the following equation for the shape factor[27]:

$$\xi = 2(l/d) \quad (6)$$

where l and d are length and thickness of the particles (l/d represents the aspect ratio of the particles). However, for our composites (containing randomly oriented particles), the value of ξ can be back-calculated through fitting the Halpin-Tsai equation with experimental data.

EXPERIMENTAL

Materials

Maleic anhydride grafted polyethylene (Epolene C-26) with an average molecular weight of 65,000 g mol^{-1}, acid number of 8 (mg KOH g^{-1}) and flow index of 8 g/10 min (190°C and 2.16 kg) was supplied by Westlake Chemical Corporation. GTR with acetone extract of 8% was obtained from Royal Mat inc. Canada. GTR particles (produced by ambient grinding method) were sieved to keep only particles between 50 and 300 μm (weighted average particle size of around 200 μm and weighted standard deviation of 46 μm). Talc powder, Stellar 410, was supplied from Luzenac America (now Imerys) and had an average particle size of 10 μm. Wood flour was a blend of sawdust from different softwood species and was kindly supplied by the Department of Wood and Forest Sciences of Université Laval, Canada. The wood flour was sieved to keep only particles smaller than 250 μm with a weighted average particle size of 170 μm (weighted standard deviation of 34 μm).

Compounding

All the samples were compounded using a co-rotating twin-screw extruder, Leistritz ZSE-27, with an L/D ratio of 40 and 10 heating zones. MAPE and GTR were fed to the first zone of the extruder (main feeder), while the rigid particles (talc and wood) were introduced in the fourth zone through a side-stuffer. The screw speed was set at 120 rpm to give a total flow rate of 4 kg h^{-1} for all samples. The temperature profile was constant along the screw at 180°C to minimize material degradation (thermo-oxidation).

For MAPE/GTR/particle compounds, the ratio of MAPE/GTR was constant at 50/50 (on a volume basis), while volume fraction of rigid particles varied between 0 and 20 vol %. For example, a composite containing 15 vol % of talc had a formulation

of MAPE/GTR/talc of 42.5/42.5/15 vol %. The extruded composites were then cooled in a water bath and pelletized at the die (5.9 mm in diameter) exit. The compounds were then compression molded in a laboratory Carver press at 200°C to form rectangular plates. The samples were first preheated for 5 min and pressed for another 5 min in molds of 115 × 115 × 2.5 mm^3 and 115 × 115 × 6 mm^3 (to prepare specimens for compression set test) under a load of 1.3 MPa.

Scanning Electron Microscopy (SEM)

Scanning electron micrographs (SEM) were used to study the morphology of the compounds and to evaluate the quality of surface adhesion between the phases. The samples were first fractured in liquid nitrogen and the surfaces were coated with a gold/palladium alloy. Then, a JEOL model JSM-840A scanning electron microscope was used to take micrographs at different magnifications at a voltage of 15 kV.

Tensile Test

Dog bone samples were cut from the compression molded plates. The tests were performed at a crosshead speed of 100 mm/min on an Instron model 5565 with a 500 N load cell at room temperature (23°C). The data reported are Young's modulus (E), tensile strength (σ), and tensile strain at break (ε_b). Each composition was tested with a minimum of five specimens to get an average. Standard deviations were less than 10% in all cases.

Compression Set

Compression sets of the thermoplastic elastomers were determined according to ASTM D395. Specimens, 13 mm in diameter and 6 mm in thickness, were cut from the compression-molded plates. The samples were first compressed to 85% of their original thickness (specimens were placed between two metal plates and the gap was controlled using screws) and then heated at 70°C for 24 h in an oven. Final thicknesses of the specimens were measured after recovering at room conditions for 30 min. Compression set values were calculated as follows:

$$\text{Compression Set} = (t_{\text{original}} - t_{\text{final}})/(t_{\text{original}} - t_{\text{compressed}}) \quad (7)$$

where t_{original}, t_{final}, and $t_{\text{compressed}}$ are the initial thickness of the sample, final thickness of the recovered sample and thickness of the compressed sample, respectively.

Each composition was tested with a minimum of three specimens to get an average. Standard deviations were less than 10% in all cases.

Thermogravimetric Analysis (TGA)

Thermal stability of the compounds was investigated with a TGA Q5000 IR (TA Instruments) at a heating rate of 10°C min^{-1} from 50 to 700°C. The tests were performed in both air and nitrogen atmospheres to evaluate the effects of oxidation on compounds stability.

Density and Hardness Measurements

Density was obtained by a gas pycnometer, ULTRAPYC 1200e, from Quantachrome Instruments, using nitrogen. Hardness (shore A) data was determined by a PTC Instruments Model 307L (ASTM D2240). In both tests, the data reported are the average of five measurements, while standard deviation was less than 5%.

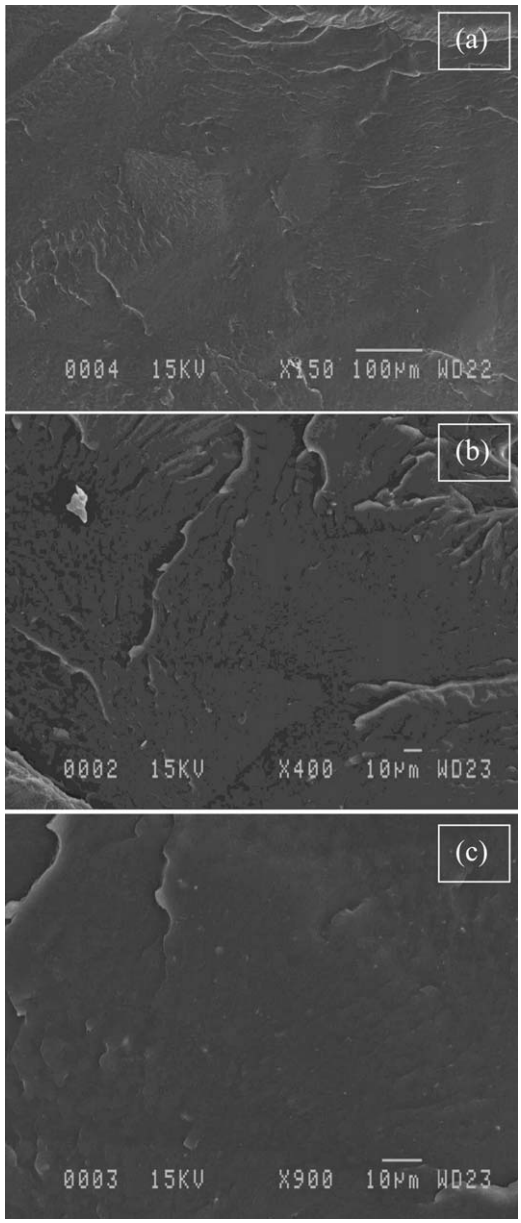

Figure 1. SEM micrographs of cryogenically fractured surfaces of MAPE/GTR (50/50).

RESULTS AND DISCUSSION

Morphological Observations

Figures 1 and 2 present SEM micrographs from cryogenically fractured surfaces of MAPE/GTR and MAPE/GTR/rigid particle compounds, respectively. The fact that no GTR particles can be detected on the surfaces of fractured samples (in Figure 1) proves that MAPE and GTR have a good level of compatibility. However, it is shown in Figure 2 that in the case of particle/matrix interactions, different behaviors can be observed. As shown in Figure 2(a,b), the level of adhesion between the MAPE matrix and talc powder was poor. Figure 2 shows that surfaces of talc particles were completely clean (of matrix) and

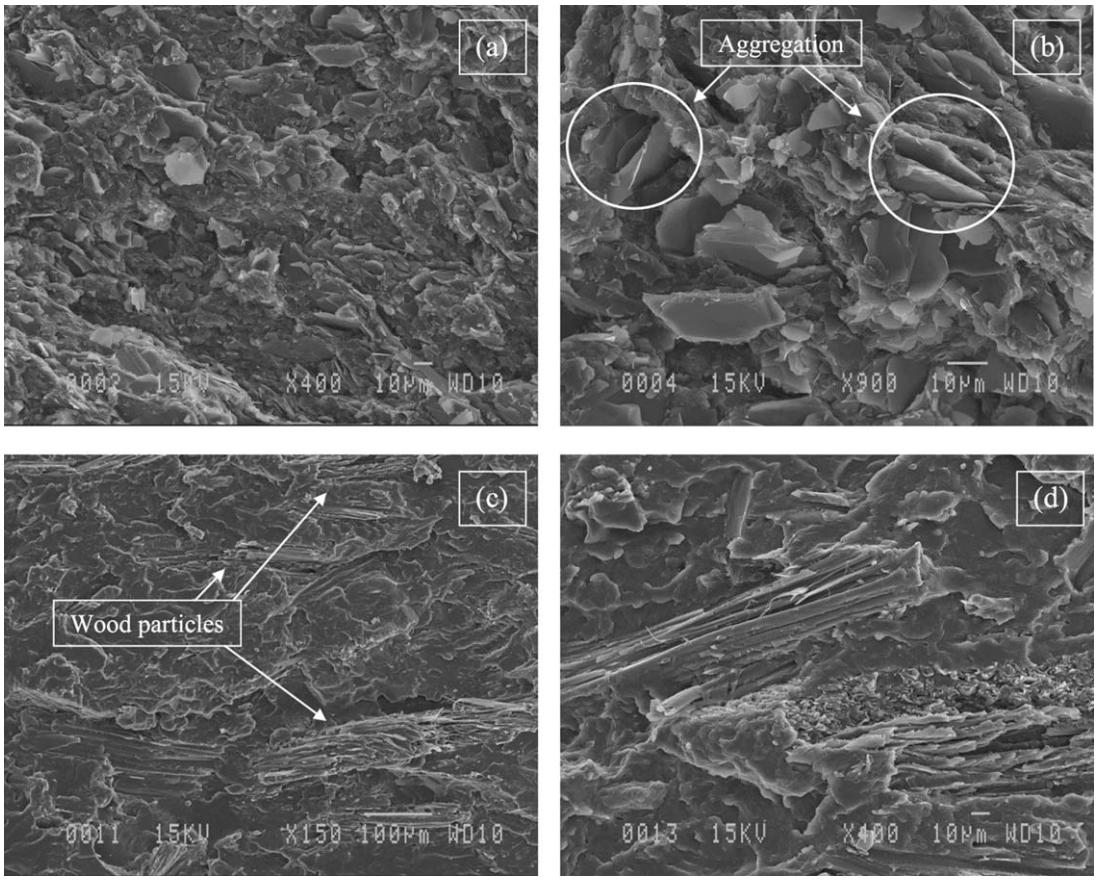

Figure 2. SEM micrographs of cryogenically fractured surfaces of composites based on MAPE/GTR (50/50) containing 20 vol % of (a,b) talc and (c,d) wood flour.

the particles can easily be spotted on the fractured surface. This observation suggests that crack propagation can occur through the weak interface. In case of compounds with good compatibility between particles and matrix, strong interface causes crack propagation to go through the matrix phase which makes particles difficult to spot. It is also shown in Figure 2(b) that the level of dispersion of talc particles in MAPE is low as signs of aggregation can be clearly seen. Wood flour, on the other hand, is remarkably well bonded to the matrix (MAPE). Figure 2(c,d) reveal that there are no gaps between wood particles and matrix. It is also seen that wood particles are broken due to the applied load. Such observations suggest that failure did not occur at the interface, indicating efficient load transfer from the matrix to the particles. High adhesion between wood flour and thermoplastic matrix is ascribed to chemical bonds between anhydride groups from MAPE and hydroxyl groups on the surface of wood particles.[28–31]

Tensile Properties

Figure 3 shows a comparison of the experimental and theoretical (calculated by eq. (1)) values for the elastic modulus of MAPE/GTR compounds. The values of parameters A and B were calculated using eqs. (2) and (3) ($A = 0.67$, $B = -1.42$). It is observed in Figure 3 that the elastic modulus of MAPE/GTR strongly decreases with GTR concentration. For example, the elastic modulus of MAPE decreased by around 31% after adding only 16% of GTR. This reduction is ascribed to the lower modulus of rubbers in comparison with thermoplastics. Figure 3 also shows that the predicted values for the elastic moduli of MAPE/GTR compounds are in good agreement with the experimental results with less than 10% deviation from the experimental values.

Figure 4 shows the experimental values of elastic modulus of MAPE/GTR compounds (E_c) reinforced with talc and wood

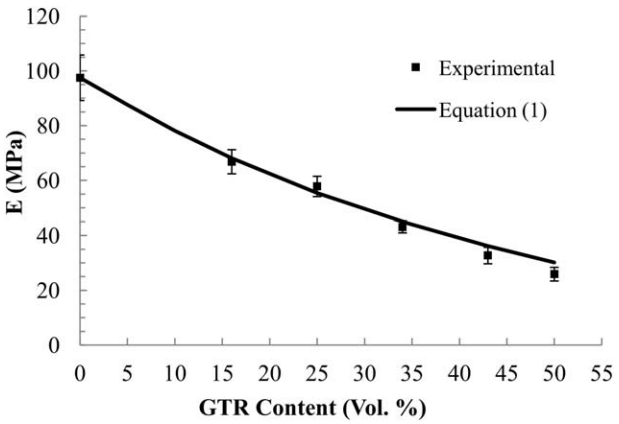

Figure 3. Experimental and predicted values for the elastic modulus of MAPE/GTR.

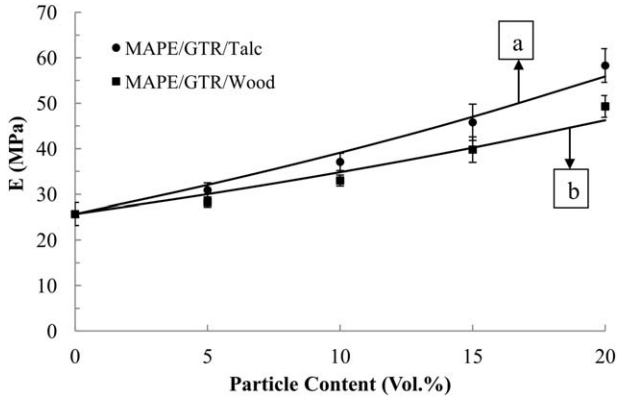

Figure 4. Experimental values for the elastic moduli with the predictions of the Halpin-Tsai equation for talc (line a) and wood flour (line b) composites.

flour. Increasing concentration of both particles increased the modulus of MAPE/GTR substantially. It is also shown in Figure 4 that for the range of concentration tested, the elastic moduli of talc-filled composites are higher compared with samples containing the same concentration of wood flour. For instance, at 20 vol % flour, the moduli are 127 and 92% higher than the MAPE/GTR (50/50) matrix for talc and wood, respectively. Higher modulus of talc-filled compounds compared to wood flour can be ascribed to higher aspect ratio of talc (average aspect ratio = 18) platelets compared with wood particles (average aspect ratio = 7) in the composites. It is also notable that average particle size of talc powder was smaller than wood flour (10 μm for talc powder compared with 170 μm for wood flour). Fu et al., however, reported that for this range of sizes, elastic moduli of particulate composites are insensitive to particle size.[34] They also concluded that only when the particle size decreases to a critical value (such as 30 nm), significant changes in modulus of composites can be observed due to changes in particle size.

Figure 4 also shows the fitting of experimental elastic moduli of talc- and wood-filled composites using the Halpin-Tsai equation. The values for effective shape factors (ξ) were calculated for MAPE/GTR/talc ($\xi = 3.7$) and MAPE/GTR/wood ($\xi = 2.2$) composites. It is shown in Figure 4 that, using these values for ξ, the elastic moduli can be predicted with high accuracy as deviations from experimental data are less than 5%.

Interesting conclusions can be drawn after comparing the calculated values of the effective shape factors of each particle with the values provided by eq. (6). It is shown that the values are much lower which is mostly due to the fact that our particles are randomly oriented. Furthermore, according to eq. (6), the shape factor for talc (ideal conditions) is 157% higher than wood (shape factors are 36 for talc compared with 14 for wood). Calculated values (by curve fitting), on the other hand, show that effective shape factor of talc is only 68% higher than that of wood. This observation is ascribed to lower compatibility between particles and matrix in talc-filled composites compared with wood-filled composites (as shown Figure 2). Presence of talc aggregates [Figure 2(b)] decreases the effective shape factor and the number of particles in MAPE/GTR/talc composites. The aforementioned results suggest that elastic modulus of particulate composites depend strongly on both aspect ratio and level of particle dispersion in the matrix.

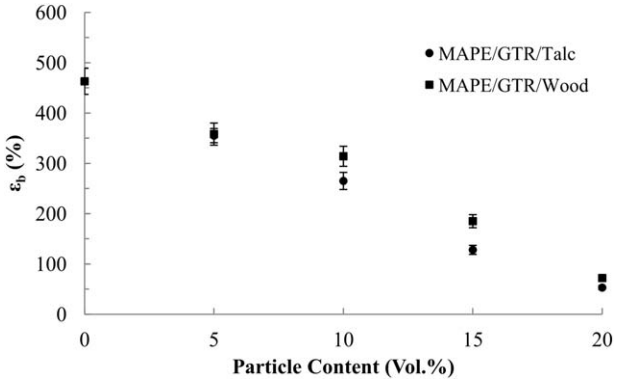

Figure 5. Tensile strain at break of MAPE/GTR/particle composites.

Figure 5 shows the effects of particle addition on the tensile strain at break of the composites. It is observed that inclusion of both talc and wood particles led to significant reduction in MAPE/GTR deformability. For instance, inclusion of only 5% of each particle reduced strain at break by almost 23%. Loss of deformability can be ascribed to several causes. In case of compounds with high compatibility between particles and matrix (i.e., wood-filled compounds), low deformability of particles leads to lower strain at break of compound in comparison with MAPE/GTR thermoplastic elastomer. In case of compounds with low compatibility between matrix and particles (i.e., talc-filled compounds) the following causes can decrease deformability: (i) possible interactions between the particles (aggregation) and (ii) stress concentration on the surface of the particles. According to the literature, presence of gaps and voids at the interface (between particle and matrix) increases the level of stress concentration which eventually causes the system to break more easily under tensile load.[33] Aggregation of particles is also reported to reduce the deformability of composites.[34]

Effect of particles on MAPE/GTR tensile strength is presented in Figure 6. It is shown that tensile strength also decreased after adding particles. Tensile strengths of composites containing 15% of talc and wood were 16 and 15% lower than MAPE/GTR,

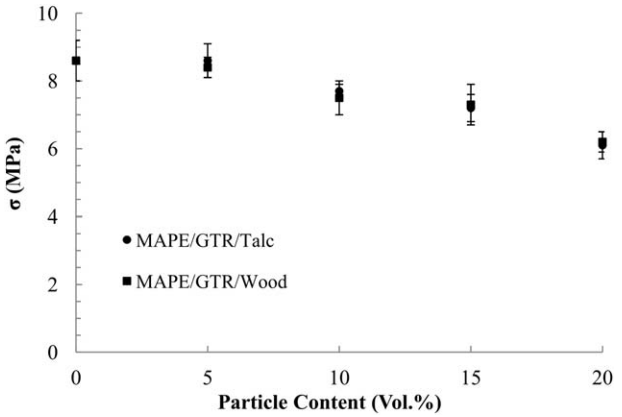

Figure 6. Tensile strength of MAPE/GTR/particle composites.

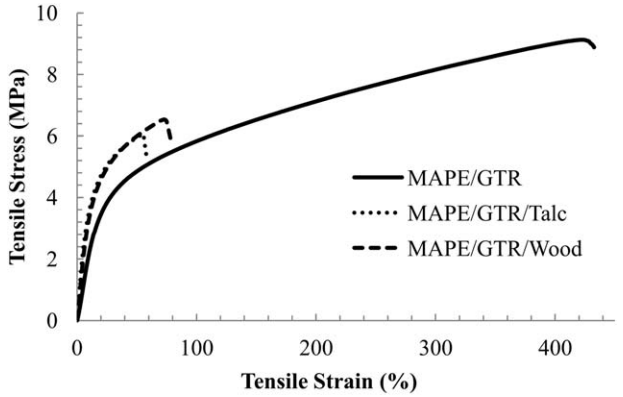

Figure 7. Stress–strain curves of MAPE/GTR:50/50 along with composites containing 20% rigid particles.

respectively. Reduction in tensile strength is related to premature failure caused by stress concentration on particle surfaces.

Figure 7 presents typical stress–strain curves from tensile test on MAPE/GTR compounds along with composites containing 20% talc and wood flour to provide more insight about their behaviors.

Compression Set

Compression set represents the elastic recovery of elastomers or thermoplastic elastomers. It is shown in Figure 8 that adding particles (both talc and wood) led to higher compression set (reduction in elastic recovery) of the composites. This behavior is due to the fact that samples with higher concentrations of rigid particles have lower rubber content leading to lower elastic recovery. For instance, samples with 20% of particles contain 40% of GTR compared to MAPE/GTR blend with 50% GTR. The authors had previously shown that a decrease in overall waste rubber content in MAPE/GTR compounds increases compression set (decreases elasticity).[16]

Thermogravimetric Analysis

Results of thermogravimetric analysis (TGA), in air and nitrogen atmospheres, are presented in Table I in terms of maximum decomposition temperature ($T_{max.\ dec}$) and T_{10} (the temperature for 10% mass loss). $T_{max.\ dec}$ represents the temperature at which the rate of thermal decomposition of the sample is at its peak. It is shown in Table I that a reduction in composite's stability occurred after increasing wood content. For instance, T_{10} of MAPE/GTR in nitrogen decreased from 334°C to 293°C after adding 20% of wood flour. However, the results suggest that $T_{max.dec.}$ did not change as significantly after inclusion of wood flour. This observation can be linked to low concentration of wood compared with MAPE and GTR.

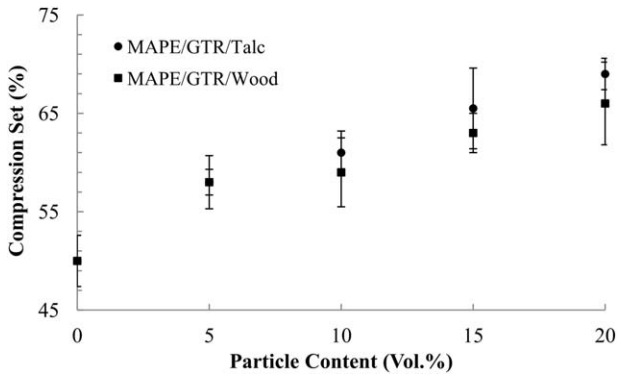

Figure 8. Compression set of MAPE/GTR/particle composites.

Table I. Thermal Degradation Results for All Samples in Air and Nitrogen Atmospheres

	$T_{max.\ dec.}$ (°C)		T_{10} (°C)	
Sample	in N_2	in air	in N_2	in air
MAPE	471	462	418	394
GTR	417	341	322	301
Wood flour	317	304	252	250
Talc	>600	>600	>600	>600
MAPE/GTR:50/50	463	452	338	336
MAPE/GTR/Talc(5)	459	450	356	352
MAPE/GTR/Talc(10)	463	452	363	357
MAPE/GTR/Talc(15)	460	449	371	362
MAPE/GTR/Talc(20)	458	451	379	368
MAPE/GTR/Wood(5)	465	455	328	309
MAPE/GTR/Wood(10)	462	449	307	301
MAPE/GTR/Wood(15)	460	447	305	293
MAPE/GTR/Wood(20)	457	445	293	288

Inclusion of talc particles did not produce significant changes in $T_{max.dec.}$ of MAPE/GTR either. On the other hand, it is observed that T_{10} of composites increased significantly after inclusion of talc. This increase is due to two reasons: (i) increased thermal stability of compounds due to higher thermal stability of talc compared with other components and (ii) the fact that degradation occurs only in MAPE/GTR compound (not in talc), while the 10% reduction in sample weight is always measured regarding to the whole composite (including weight of talc).

As expected, presence of oxygen in air decreased the stability of all composites. Maximum decomposition rate was observed at 463 and 452°C for MAPE/GTR compound in nitrogen and air, respectively (see Supporting Information for DTG curves). TGA

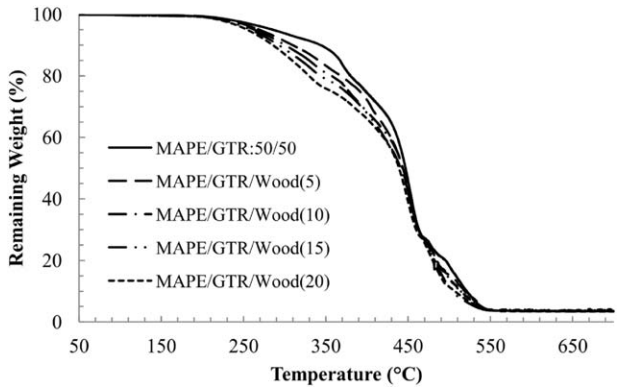

Figure 9. TGA plots for MAPE/GTR compounds with different concentrations of wood flour (in air).

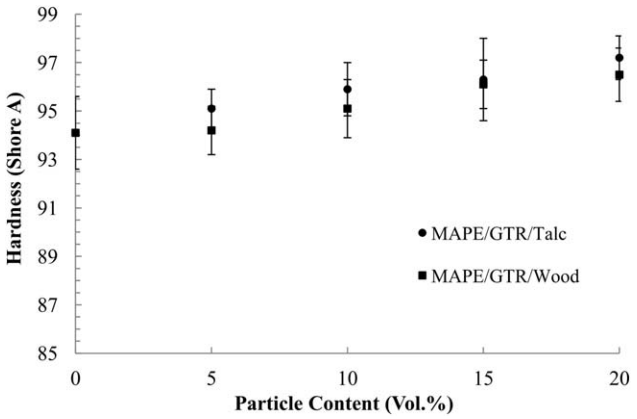

Figure 10. Hardness of composites with different reinforcements.

plots of MAPE/GTR/wood composites in air are presented in Figure 9 and show that ultimate weight losses are less than 100%. This is due to the presence of impurities in the GTR phase which could affect the properties of the blends.

Density and Hardness Measurement

From density measurements, it was observed that densities of composites filled with wood flour were considerably lower than composites with talc (see Supporting Information). For instance, densities of composites containing 20% of wood and talc were 1.13 and 1.44 g cm^{-3}, respectively. This is due to the lower density of wood (1.4 g cm^{-3}) compared with talc (2.9 g cm^{-3}). Lower density of wood flour-filled composites causes them to have comparable specific mechanical properties (property/specific gravity) with talc-filled composites.

Figure 10 shows that the addition of particles led to a slight increase in hardness. Although the hardness of both composites (talc- and wood-filled composites) was close, it is shown that hardness of composites containing talc powder was slightly higher than wood-filled composites. For instance, hardness of composites containing 20% of talc and wood were 97.2 and 96.5 shore A, respectively.

CONCLUSIONS

The effects of particulate reinforcements (talc and wood flour) on the properties of MAPE/GTR thermoplastic elastomers were investigated. SEM results revealed that MAPE had a good level of adhesion with wood flour. However, composites containing talc had low particle-matrix compatibility and signs of talc aggregation were observed. Adding both types of particles was shown to increase tensile modulus of the thermoplastic elastomers significantly. For instance, MAPE/GTR/talc and MAPE/GTR/wood composites containing 20% of reinforcement were respectively 127 and 92% stiffer than the MAPE/GTR (50/50) compound. Modeling of the elastic modulus of MAPE/GTR thermoplastic elastomers and MAPE/GTR/particle composites was successfully performed using Kerner and Halpin-Tsai equations, respectively. Fitting of the experimental data using the Halpin-Tsai equation showed that the elastic moduli depend strongly on both aspect ratio and level of dispersion of the reinforcements in the matrix. Although inclusion of both particles led to reduction in strain at break and tensile strength of MAPE/GTR compounds, it was shown that even for composites containing up to 15% of particles, the strain at breaks were higher than 100%.

Increasing particle concentration also resulted in lower elastic recovery of MAPE/GTR compounds. Compression set of the thermoplastic elastomers increased from 50 to 69% after adding 20% of talc powder to MAPE/GTR. Composites with higher compatibility between the particles and the MAPE matrix showed stronger elastic behavior. Thermal stability of compounds with talc powder was not affected by particle content, while inclusion of wood flour decreased thermal stability slightly. For instance, T_{10} of MAPE/GTR in nitrogen decreased from 334°C to 293°C after adding 20% of wood flour. The composites showed lower thermal stability in air compared to nitrogen which is linked to oxidation. Adding the particles led to increased density and hardness of MAPE/GTR, but both properties were higher for talc-filled composites.

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