The effect of recycled Natural Rubber Glove (rRG) Plasticizers to Deflection and Flexural Strength Properties of PP/MMt/rRG Smart Composites and Its Inflammability

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Abstract. Had been synthesized PP/rRG/MMt+ZB smart material composite in solution reactive processes with various rRG concentration. The addition of rRG plasticizers will improve the deflection properties and increase the filler capacity MMt loading to reach the optimum concentration. The addition of 3% rRG is capable of loading filler capacity MMt to 23% as the optimum condition. At the optimum conditions it can increase the deflection (Defl) and flexural strength (FS) up to 16% and 15% respectively compared to that of the composites without rRG. The rRG plasticizer serves as a bio-compatibilizer that can reduce surface tension of the mixture and leads to decrease the Defl, follow by the increase of loading filler capacity and well interaction finally can increase the FS properties. The increase of loading filler MMt up to 23% can also improve the inflammability of the composites. Time to Ignition (TTI) increase by 5% and Burning Rate (BR) decrease by 4.5% compared to that of the composites which is containing MMt 20% without rRG.

1. Introduction
The availability of metals in nature is increasingly depleted while the production of automotive that uses metal as its raw material production is increasing by 3.2% every year [1]. Therefore its need alternative material of metal substitute which have strong, light and flame resistance properties, that is composite. The use of 35% lightweight composite on automotive can save 12-20% fuel usage [2]. The composite is very suitable to be applied in automotive components such as upholstery, door panel, interior and car body [3]. Increased mechanical properties and flame resistance of a composite can be accomplished by the addition of clays such as montmorillonite (MMt) [4]. The type of MMt clay is abundant in Indonesia and can be used as a composite filler material. Lee made PP/WF/MMt composite with PP-g-MA coupling compound internally mixer producing composite reported to have high in tensile strength (TS) and melting temperature [5].

Elastomeric composite engineering is one of them with the addition of a composite-like plasticizer. Wang made the PP/EPDM/SGF composite to produce composites with high elasticity and mechanical

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properties [6]. Xie and others make composite PP/styrene-butadienerubber (SBR)/SGF produce composites with high elasticity and mechanical properties [7]. Balakrishnan makes composite PP/epoxydized natural rubber (ENR)/OMMt produces composites with high elasticity and mechanical properties [8]. Ismail and Suryadiansyah made a composite PP/recycle natural rubber (rNR) reported to have high elasticity and mechanical properties compared to PP [9]. Riyajan make composite PS/recycle rubber glove (rRG)/compostable leaf cane produced with elasticity and high mechanical properties compared to starting materials [10]. The objective of the research is to study the effect of adding rRG plasticizer to rPP/DVB/PP-AA/MMt+ZB composites to deflection, flexural strength and its inflammability properties.

2. Experimental

2.1 Materials

Recycled polypropylene (rPP) was collected from cup recycled plastics packaging of mineral water in Surakarta, Indonesia. The rPP was washed and then chopped approximately in 5 mm x 2 mm. The montmorillonite (MMt) clay was obtained from Soka tile Kebumen Indonesia. The MMt prepared by crushed and sieved with a 200 mesh sieve and calcined at 800°C for an hour. The recycle natural Rubber Glove (rRG) was collected from Surakarta, Indonesia. Preparation of rRG by decrosslink processed using two roll mill and then crushed and sieve with a 40 mesh. All chemicals were pro-analysis without any purification. The acrylic acid (AA) purchased from Schuchai, divinyl benzene (DVB) purchased from Aldrich, xylene derived from PT. Brataco in Surakarta, Indonesia. While benzoyl peroxide (BPO) and zinc borate (ZB) purchased from E.Merck.

2.2 Composites manufacturing process

Fragments of rPP and a number of rRG are mixed in the shaker and then inserted in a chamber containing boiled xylene. The system is equipped with an oil bath heated at 135 °C., thermometer, mechanical stirrer, and N2 gas flow of 0.25 Barr/hr so that the rPP melts and then BPO initiator were added. Furthermore, PP-AA, DVB and MMt are inserted to the mixture compounds and the heating reactive process was carried out for 1 hour. The composite making composition is in accordance with the formula shown by Table 1.

| Formulas | Code | Composition (%) | rPP | rRG | DVB | PP-AA | MMt |
|----------|------|-----------------|-----|-----|-----|-------|-----|
| rPP      | MF0  | 100 0 0 0 0     |     |     |     |       |     |
| rPP/DVB/PP-AA/MMt20%+ZB | MF1  | 65 0.1 15 20    |     |     |     |       |     |
| rPP/DVB/PP-AA/0%rRG/MMt23+ZB | MF2  | 65 0.1 15 23    |     |     |     |       |     |
| rPP/DVB/PPAA/1%rRG/MMt23+ZB | MF3  | 60 1 15 23     |     |     |     |       |     |
| rPP/DVB/PP-AA/3%rRG/MMt23+ZB | MF4  | 60 3 15 23     |     |     |     |       |     |
| rPP/DVB/PPAA/5%rRG/MMt23+ZB | MF5  | 60 5 15 23     |     |     |     |       |     |
| rPP/DVB/PP-AA/7%rRG/MMt23+ZB | MF6  | 60 7 15 23     |     |     |     |       |     |

2.3 Testing of materials

The characterization of a composite functional group was done by the FTIR Prestige 21 Shimadzu. The testing of Flexural Strength (FS), Deflection, and Bending Elasticity (BE) was carried out using universal testing machine (UTM) model JTM-UTS510 by following ASTM D790 with a three point bending method. The testing of flame resilience of composites in accordance with ASTM D365 to determine the value of time to ignition (TTI) and burning rate (BR).
3. Results and Discussion

3.1 FTIR Analysis Functional Groups of Composites

The result of FTIR characterization on MMT was obtained Si-O at 1084 cm\(^{-1}\), Al-O-Si absorption at 686 cm\(^{-1}\), Si-O-Si uptake at 480 cm\(^{-1}\) and –OH absorption at 3447 cm\(^{-1}\). In the PP/DVB/PP-AA/rRG composites, the typical uptake of rPP is the >C-H bond of methyne group at 1723 cm\(^{-1}\) and vibration stretching of carbonyl group (=C=O) at 1718 cm\(^{-1}\) indicates carbonyl acid of PP-AA [4,11,12,13]. In composites of PP/DVB/PP-AA/rRG/MMt+ZB, functional group of Al-O-Si can be indicated as the absorption of Al-OH at 867 cm\(^{-1}\), absorption of Si-O-Si at 1088 cm\(^{-1}\), and Si-O-Si uptake at 486 cm\(^{-1}\) indicates the presence of MMT with the main content of SiO\(_2\) and Al\(_2\)O\(_3\) [14,15]. The broad vibration stretching of O-H bond at 3460 cm\(^{-1}\) that indicate hydrogen bond of >Al-OH of MMT. This is supported by the previous research [14,16,17]. From the results of FTIR spectra analyzes mentioned above, the composites using raw materials of rPP, rRG, PP-g-AA, ZB and MMT have been clearly indicated.

![FTIR spectra of MMT, PP/DVB/PP-AA/rRG, PP/DVB/PP-AA/rRG/MMt+ZB](image)

**Figure 1.** FTIR spectra of (a) MMT, (b) PP/DVB/PP-AA/rRG, (c) PP/DVB/PP-AA/rRG/MMt+ZB

3.2 Mechanical Properties of Composites

The mechanical properties of composites are presented in Figure 1-6. In Figure 2-3 show the effect of plasticizers rRG to strength properties of PP/DVB/PP-AA/rRG/MMt+ZB (MF2-MF6 containing 23% MMT with various concentration of rRG). The optimum FS of PP/DVB/PP-AA/MMt+ZB composites (MF1, without rRG plasticizer) have content 20% MMT was 39.32 MPa, however, when the contain of MMT added to 23% (MF2) the FS decreased to 5% compared to that of MF1. In the addition of 1% and 3% of rRG plasticizer (MF3 and MF4) containing 23% MMT can increase 8% and 15% of FS compared to MF1, respectively. This happens because of the addition of a rRG plasticizers on the composite will reduce the surface tension of the mixture, it causes the mixture to become softer, follows by the increase in elasticity and consequence the value of the deflection will increase. This softer material properties can receive a larger MMT filler capacity of up to 23% under optimum conditions. Increased the MMT filler automatically functions as an reinforcement thus increasing the FS value of composite [4,13,18]. While the addition of 5% and 7% of rRG (MF5 and MF6) containing...
23% MMt decreased 11% and 19% of FS compared to MF1. The decrease in mechanical properties are due to the greater content of plasticizer material resulting in too soft material and finally lead to lower mechanical properties.

Figure 2. Flexural Strength (FS) of the composites (MPa)

Figure 3. Comparison of increasing FS (%) to that of MF1

Figure 4. Deflection of the composites (mm)

Figure 5. Comparison of increasing deflection (%) to that of MF1

Figure 6. Bending Elasticity (BE) values of the composites (MPa)

Figure 7. Comparison of increasing BE (%) to that of MF1

Figure 4 and 5 show the deflection (Defl) value of composites MF1-MF6. The Defl value of MF1 without rRG plasticizer and have content 20% MMt was 11.9 mm, however, when the MMt filler added to 23% of MMt (MF2), the Defl. value decreased 7% compared to that of MF1. This is because, the addition of filler above the optimum capacity conditions will result in interaction between raw material does not occur well, consequence in the brittle properties of the composite. This is supported by Othman which is reported that the higher filler content will decrease the value of strain (elasticity) which eventually decrease the deflection value [19]. The composites (MF2-MF6) which have content 1%, 3%, 5%, and 7% of rRG plasticizer respectively and contain 23% of MMt filler, have the Defl. value increase to 8%, 16%, 18%, 24% respectively compared to that of MF1. This increase is supported by the previous work, that the higher rubber will increase the value of the strain resulting in an increase in deflection [9,10]. Figure 6 display the bending elasticity (BE) and Figure 7 display its comparison to that of MF1. The stiffness properties of the material will decrease as elasticity increases. Likewise, what happens in Figure 6 and 4. The BE values of the composites in figure 6
shows a decreasing profile when Defl. values in Figure 4 increases. The BE composite value of MF1 without plasticizer is 2753.5 whereas MF2 is 2804.8 MPa. Its because the Defl value in MF2 is lower than MF1, due to the amount of filler MMt on HM2 is larger then MF1. Profile of composite EB value of MF2 to MF6, showing downward trend with respect to rRG plasticizer content go up (Defl value increase).

3.3 Flammability Properties

Figure 8-13 show the flammability of the rPP and filled composites (formulas MF1-MF6) measured by horizontal burning test following ASTM D635. The effect of addition of rRG plasticizer to time to ignition (TTI) and burning rate (BR) of rPP/DVB/PP-AA/rRG/MMt+ZB composites (MF2-MF6 which is containing 23% of MMt in various concentration of rRG) were showed in Figure 8-11.

![Figure 8. TTI of the composites sec)](image1)

![Figure 9. Comparison of increasing TTI to that of MF1 (%)](image2)

![Figure 10. BR of the composites (mm/min)](image3)

![Figure 11. Comparison of increasing BR (%)](image4)

The TTI and BR of MF1 (without rRG plasticizer) were 6.02 s and 11.58 mm/minutes. When the filler become 23% of MMt (MF2), the TTI increased 15.95% while the BR decreased 9% to that MF1. Meanwhile the 1% and 3% concentration of rRG (MF3 and MF4) with 23% of MMt, the TTI increased 10.47% and 4.98%, while the BR decreased 6.68% and 4.5% compared to MF1. At 5% and 7% concentration of rRG (MF5 and MF6), the TTI decreased 6.15% and 9.97% while the BR increased 2.73% and 7.28% compared to MF1. MMt content and ZB in the composite on the combustion process undergo endothermic reactions and produce water vapor, charcoal and humid B₂O₃. This will lowering the temperature, limiting the intake of O₂ thus inhibiting the combustion process, this is supporting by the previous work [13,20]. The highest TTI and the lowest BR show that the material has high inflammability.

The heat release (HR) is the ability of a material to release heat after burning is extinguished at a certain period of time. Thus, the percentage of heat released during that period of time can be measured by using the equation HR (%)={\[(1-T_f/T_i)\times 100\%\]}, where, T_f is the material’s temperature after the period has elapsed, and T_i is combustion temperature. In Figure 12-13 show the effect of rRG addition to that %HR of composites. The %HR of MF1 without rRG plasticizer, with 20% of MMt filler was 66.55%, however, when the filler became 23% the %HR increased 5.09% compare to
MF1. This is supported by Du and Othman who stated that the higher the clay concentration in the composite the flame retardant properties are increasing and the composite is not flammable [19,21].

![Figure 12. %HR of the composites (%)](image1)

![Figure 13. Comparison of Increasing %HR](image2)

Delhom reported that in the event of burning clay will form a charcoal that can cover the composite surface so as to reduce the contact between the composite with oxygen causing the burning process is inhibited [22]. In addition, the presence of clays in the polymer matrix causes an endothermic reaction resulting in reduced combustion temperatures and burning being impeded [23]. In addition of rRG plasticizer can increase the loading capacity resulting the high inflammability. The %HR of MF3 and MF4 can increase the HR% by 2.83% and 1.50% against MF1. However, composites MF5 and MF6 decreased %HR by 1.8% and 6.27% respectively compared to MF1. The decrease is due to the higher rRG concentration. This is in accordance with Balakrishnan who state the value of 10% mass degradation temperature that the higher the thermal endurance rubber content decreases [8]. This is also supporting by the previous work Patra and Suharty [24,25]. Material that has a high % heat release also indicates a high fuel resistance.

4. Conclusion
Addition of 3% rRG plasticizer of rPP/DVB/PP-AA/MMt+ZB is able to increase 15% filler capacity on composite. At optimum condition without plasticizer the filler capacity of MMt is only 20% but with addition of 3% pemplastis obtained optimum condition of filler loading capacity increased to 23%. The addition of 3% rRG pemplastis capable of increasing filler loading capacity, it will automatically improve the mechanical properties and combustion resistance of the composite. The mechanical properties of rPP/DVB/PP-AA/3% rRG/23% MMt+ZB new smart composite is as follows. The FS value is 45.08 MPa, deflection is 13.8 mm and bending elasticity (BE) is 2722 MPa. The effect of addition 3% rRG plasticizer can increase the FS by 15%, increase the deflection by 16% and decrease the BE by 1% compared to that of MFI composite without rRG plasticizer. The inflammability properties value of new smart material is as follows. The TTI is 6.32 seconds, BR is 11.04 mm/min, and HR is 67.55%. The effect of addition 3% rRG plastics can increase TTI to 4.98%, decrease BR to 4.50% and increase HR to 1.5% compared to that of MFI composites without rRG plasticizer.

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