Smart Natural Fiber Reinforced Plastic (NFRP) Composites Based On Recycled Polypropylene in The Presence Kaolin

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Abstract. Composites contain double filler material which act as reinforcement and flame retardants of recycled polypropylene (rPP)/kaolin(Kao)/palm oil empty bunch fiber (PEBF) have been successfully prepared. The composites were synthesized through reactive solution method, using coupling agent PP-g- AA and compatibilizer DVB. The effect of double filler [Kao/PEBF] were investigated flexural strength (FS), inflammability, and morphology. Mechanical testing result in accordance to ASTM D790, the FS of rPP/DVB/PP-g-AA/Kao+ZB/PEBF composite was 48% higher than that of rPP matrix. Moreover, flexural modulus (FM) was significantly improved by 56% as compared to that of rPP matrix. The scanning electron images (SEM) shown good dispersion of [Ka/PEBF] and good filler-matrix interaction. The inflammability testing result which is tested using ASTM D635, showed that the flame resistance of rPP/DVB/PP-g-AA/Kao+ZB/PEBF composite was improve by increasing of time to ignition (TTI) about 857% and burning rate (BR) decreasing to 66% compared to the raw material rPP matrix. In the same time, the addition of 20% (w/w) PEBF as a second filler to form rPP/DVB/PP-g-AA/Kao+ZB/PEBF composites (F5) is able to increase: the FS by 17.5%, the FM by 19%, the TTI by 7.6% and the BR by 3.7% compared to the composite without PEBF (F2).

1. Introduction

Today, the automotive production is increasing, however, availability of metal as raw material for the automotive is also depleting [1]. In addition, the fuel oil is a material supporting automotive operations, its availability is also going decrease significantly [2]. This problems can be overcome by the use of light weight automotive. The light weight automotive can be created by partially replacing metal in automotive by composite materials [3]. Composite is a very interesting material by nature to produce new materials with new properties as required. Manufacture of high mechanical properties composite can be done by adding a clay or fiber as reinforcement in polymer matrix. One alternative development composites is by creating smart composite Natural Fiber Reinforced Plastic (NFRP) which has advantages such as light in weight, strong and biodegradable [4,5]. Polypropylene is a plastic used widely in various applications such as electronic devices, transportation and packaging [6,7]. Polypropylene in the packaging of mineral water waste overflow and pollute the environment, it can be used as a polymer matrix [8,9]. The use of fiber as reinforcing filler for plastic composites have been used extensively. Maulida [10] synthesized biocomposite of acrylonitrile butadiene styrene (ABS) with PEBF (palm oil empty fruit bunch fiber) was obtained composites have high mechanical properties. Wirjosentono [11] synthesized bio-composites of polypropylene with PEBF was obtained composites
have high mechanical properties. Other reinforcing material that has been used by previous researchers is clays [12-15]. Any clay other than as a reinforcing material can also act as flame retardants. Shehata [16] synthesized a composite of polypropylene and kaolin clay to produce a composite that has high flame resistance. Du [17] make a composite of polypropylene and halloysite clays produce a composite that has high flame resistance.

In this research will synthesized: (1) rPP/DVB/PP-g-AA/Kao+ZB composite by using the rPP as a matrix, divinylbenzene (DVB) as a crosslinker and PP-g-AA as a coupling agent and the kaolin (Kao) as a first reinforcement, (2) rPP/DVB/PP-g-AA/Kao+ZB/PEBF composite containing palm oil empty bunch fiber (PEBF) and Kaolin has used as a double reinforcement filler. The composites in the form double filler PEBF and Kaolin, which produces NFRP composites with mechanical properties and durability of high flame and biodegradable. GeoBioComposites are the development of green composites that fulfill the criteria to be applied in automotive components.

2. Methods
2.1 Materials
Recycled polypropylene (rPP) was collected from plastic recycling centre in Surakarta Indonesia. The rPP prepared by cleaning up the waste cup and then chopped approximately in 2 mm x 2 mm. PEBF obtained from oil palm plantations in Lampung Indonesia. The PEBF was then alkalized with 4% NaOH for one night, and washed with distilled water and then dried in an oven at 60 °C before grounded into 200 mesh sized particles. The kaolin purchased from PT Brataco in Surakarta Indonesia. It crushed and sieved with a 200 mesh sieve and calcined at temperatures of 800 °C for 1 hour. All chemicals used in this study were pro-analysis without any purification. Other chemical compounds, such as acrylic acid (AA) and divinyl benzene (DVB), xylene derived from Aldrich; while benzoyl peroxide (BPO) and zinc borate (ZB) purchased from E.Merck.

2.2 Composites manufacturing process
Formulations of rPP (F0), rPP/DVB/PP-g-AA/Kao+ZB (F2), rPP/DVB/PP-g-AA/PEBF (F3) and rPP/DVB/PP-g-AA/Kao+ZB/PEBF (F5) composites synthesis are given Table 1. Composite synthesis is reactively done by solution process using xylene, equipped with a condenser, mechanical stirrer and N2 gas. All starting materials: rPP, DVB, PP-g-AA, Kaolin, and PEBF solved in boiled xylene and mixed perfectly for 1 hour. The mixture product was then evaporated to release the xylene to form masterbatches (MB’s) composites. The MB’s was hot pressed for 20 minutes at 180 °C to produce the specimen for mechanical test [8].

| Ingredients (phr) | F0  | F1  | F2  | F3  | F4  | F5  |
|-------------------|-----|-----|-----|-----|-----|-----|
| rPP               | 100 | 80  | 80  | 80  | 80  | 80  |
| PEBF              | -   | -   | -   | 20  | 20  | 20  |
| Kaolin            | -   | 20  | 20  | -   | 20  | 20  |
| ZB                | -   | -   | 5   | -   | -   | 5   |

2.3 Testing of materials
The testing of Flexural Strength (FS) and Flexural Modulus (FM) was carried out using universal testing machine (UTM) by following ASTM D790. The testing of flame resilience of composites in accordance with ASTM D635 to determine the value of time to ignition (TTI) and burning rate (BR).

2.4 Morphology analysing
The fracture surface of specimens after FS testing were characterized using SEM-EDX JEOL JSM-6360 LA to analyse their morphology, especially on the cross section area the fracture surface.
3. Results and Discussion

3.1 Mechanical properties of composites

The mechanical properties of composites are presented in Figure 1-4. The FS and FM of rPP (F0) as raw materials was 23.61 MPa and 790 MPa. The FS of rPP/Kao+ZB (F2) enhanced 16.5% (27.5 MPa) and the FM also increased 22% (960 MPa) due to the effect of the adding 20% (w/w) kaolin as a first reinforcement. Meanwhile, the presence 20% (w/w) PEBF on rPP (F3) produce rPP/PEBF composite with 30.11 MPa of FS increased 23% and 1060 MPa of FM also increased 29% compared to rPP. In the addition of 20% (w/w) kaolin as a first reinforcement and 20% (w/w) PEBF as a second reinforcement (F5), the FS of rPP/Kao+ZB/PEBF composites become 32.3 MPa in FS and 1140 MPa in FM, or the FS increased 48% and the FM increased 56% compared to that of rPP (Figure 1, 2, 3 and 4). The increases was caused by both PEBF and kaolin act as a reinforcing material [14,18,19].

![Flexural Strength (MPa) vs Composites](image1)

![Increasing of FS (%) vs Composites](image2)

![Flexural Modulus (MPa) vs Composites](image3)

![Increasing of FM (%) vs Composites](image4)

The existence of a reinforcing material in the form of fibers within the polymer matrix serves to hold the burden that has been transferred from the matrix to the fiber. Chemically, PEBF have polar groups such as -OH are able to interact chemically with metin group of RPP through a coupling compound PP-
g-AA [7]. Furthermore, the presence of kaolin clay reinforcing material that has a layer of type 1:1 can improve the mechanical properties. It is caused by the geometry of the clay in the polymer matrix can be distributed both intercalation and exfoliation [20,21]. Judging from the chemical side, kaolin clay has a layer in the form of Si-O tetrahedral and octahedral layers of Al-OH which are capable of interacting with polar groups on a polypropylene metin through the coupling compound PP-g-AA. Maximum interaction between filler and kaolin PEBF with PP matrix will lead to increased mechanical properties [7].

3.2 Inflammability properties

To prove the inflammability of various (F0-F5) composites its performed resistance combustion testing. The method horizontal burning of ASTM D635 is used for testing composites. The result is presented by the time to ignition (TTI) and the burning rate (BR), which can be seen in Figure 5-8. The TTI and BR of rPP (F0) was 1.02 second and 19 mm/mins, and in the presences of 20% (w/w) kaolin (F2), the TTI of rPP/kaolin+ZB increased 806% (9.2 second), however, the BR decreased 48% (9.8 second) compared to rPP. The increased of TTI and the decreased of BR value its due to the effect of 20% kaolin + 5% ZB. When kaolin + ZB burned, its will form the charcoal and overlying the surface of the specimen, thus limiting the supply of oxygen. This condition can inhibit the TTI and slow down its BR. The presence of 20% (w/w) PEBF (F5) in rPP/kaolin+ZB/PEBF, the TTI increased to 857% (9.9 seconds) and the BR decreased to 66% (7.8 mm/mins) compared to that of rPP. Cellulose fibers of PEBF during the combustion process formed charcoal as a barrier that prevents the entry of oxygen at the surface. Kaolin undergo endothermic reaction which can absorb heat energy during the combustion process takes place, while ZB can release water vapor to cool the system [19,22,23]. The synergy of these three components causing the flame resistance of the composite rPP/DVB/PP-g-AA/Kao+ZB/PEBF (F5) increased significantly. The presence of 5% ZB as fire retardant additive can inhibited TTI and also slow down the BR. The ZB compound binds some water when its burnt will release the water vapour. ZB if burned is also formed B2O3 which is moist and form a stable charcoal. Water that is released by ZB during combustion can cause charcoal to form a foam that inhibits combustion [18,22,23].
The mechanical and inflammability properties values of rPP/DVB/PP-g-AA/Kao+ZB: the FS is 27.5 MPa, FM is 960 MPa, TTI is 9.2 seconds and BR is 8.1 mm/mins. The effect of mechanical and inflammability properties values by adding 20% (w/w) PEBF is as follow: the FS values increased 17.5%, the FM values increased 19%, the TTI values increased 7.6% and the BR values decreased 3.7% compared to rPP/DVB/PP-g-AA/Kao+ZB composites without PEBF.

3.3 Morphologies analysis
The fracture surface of specimens after FS testing were characterized using SEM image especially on the cross section area of the fracture surface. The morphological study carried out by compare the composite cross-section SEM image of the fracture of the specimen testing FS rPP/Kao without and with PEBF, can be seen at Figure 9 (F2) and Figure 10 (F5). The observation of SEM image F2 shown that the kaolin particles as seen at points A very well dispersed in the PP matrix. However, at point B, the result of fracturing after FS testing showed the brittle nature of the material without PEBF (F2), its mean the interaction between filler and matrix is good. Morphology of composite rPP/Kao/PEBF (F5) can be seen in the SEM image on points D and E, both filler kaolin and PEBF dispersed very well. And also at points C after FS test shown the ductile composite nature. This means there has been a very strong interaction between filler and matrix, resulting a strong mechanical properties of the composites.
4. Conclusion
The mechanical and inflammability properties values of rPP/DVB/PP-g-AA/Kao+ZB is as follow: FS is 27.5 MPa, FM is 960 MPa, TTI is 9.2 seconds and BR is 8.1 mm/mins. The effect of mechanical and inflammability properties values by adding 20% PEBF is as follow: the FS values increased 48%, the FM values increased 56%, the TTI values increased 857% and the BR values decreased 66% compared to rPP as raw material. In the same time, the effect of mechanical and inflammability properties values by adding 20% PEBF is as follow: the FS values increased 17.5%, the FM values increased 19%, the TTI values increased 7.6% and the BR values decreased 3.7% compared to rPP/DVB/PP-g-AA/Kao+ZB composites without PEBF.

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