Study on formation and mechanical properties of rice husk/natural bentonite prepared composites

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Abstract In this study, composites were made from source of virgin HDPE and rice husk/natural bentonite as combined filler and mechanical properties of composites were measured. The purpose of study is to find the best condition of the ratio of rice husk/natural bentonite as matrix and the percentage of polyethylene-graft-maleic anhydride (MAPE) as coupling agent on quality of formed composites. In order to form these composites a hot press method was performed. The composite formation process was conducted at temperature of 170 °C for half hour. After pressed and heated then composites was cooled to room temperature and finally composites were tested to know its mechanical properties of tensile strength and bending strength. The results showed that addition of natural bentonite into matrix of rice husk have increased mechanical properties of composite compared to composites prepared by using only rice husk and virgin HDPE. Percentage of added MAPE also affect values of tensile and bending strength of composites. The effect of MAPE and natural bentonite which added during formation of composites caused the increasing adhesion bond between matrix and filler and as a result also increased the mechanical properties of composites. The highest tensile strength value of 18.2 MPa was found on composite with filler rice husk/natural bentonite ratio of 70:30 with addition of 5 wt% MAPE. While the highest bending strength value of 9.2 Mpa was obtained on composite with filler rice husk/natural bentonite ratio of 70:30 and addition of 5 wt% MAPE.
Key words : natural bentonite, polyethelene, composite, MAPE and rice husk

Introduction

Nowadays, the need of wood to support human being life has been increasing rapidly with increasing human being population. Generally, people use wood for building materials or other applications. With increasing population, wood utilization has been also increasing sharply. In order to find a new material to replace the use of wood, researchers try to study and synthesize others materials. One of materials that can substitute timber or wood are composite. Composite can be produce from many materials but preparation of composites by using biomass can reduce the environmental load due to amount of biomass in our atmosphere and also produces innovative products as a substitute for wood or timber. In addition the use of bentonite that is naturally available in huge amount in soil surface would also other benefits. The advantages of biomass/bentonite based composite products, among others are low density, more economical production cost, readily available raw materials, flexible in its construction and other better properties. There are a lot of biomass as agricultural by-product in Aceh Province that has not been fully utilized such as rice husk, sawdust, bamboo fiber, straw and so forth. Actually, those biomass can be processed to be new materials by mixed with plastic in order to obtain a material that has different properties with its basic property (Roger M. R. 2007).

Biomass-based materials such as rice husk, sawdust, bamboo fiber or straw are hydrophilic, rigid and degradable after some time in enviromental condition. Those specific properties of material causes less suitable when combined with non-organic material such as polimer that is hydrophobic without addition of coupling agent. Addition of coupling agent would improve bonding interaction between matrixs and fillers as previously studied by author (Farid, M. 2012). In this study, the insertion of small amount of natural bentonite would be also performed and it is hoped that addition of coupling agent would improve adhesion bonding between matrix and filler and finally improve mechanical properties of composites. It is well known that some coupling agents can be used to produce wood polymer composites such as organic coupling agent, inorganic and organic-inorganic coupling agent. To enhance bonding interaction between matrix with an inorganic filler, some previous researchers have used various types of clay beside using coupling agents (Bledzkia K., et al. 1998, Ali H. A., et al. 2012, Mohanty S., et al. 2006, John Z. L., et al. 2005, Farid M. 2013, Ravindra C. R., et al. 2010). In this study the using of maleic anhydride based polymers of maleic anhydride polyethylene type (MAPE) as coupling agent and natural bentonite as filler was investigated. The applicability of maleic anhydride based polymers coupling agent provide the best results where MAPE can increase effectively phase interaction bonding between a polar filler and a non-polar polyethylene. The aim of study is to find the best condition of the ratio of rice husk/natural bentonite as matrix and the percentage of polyethylene-graft-maleic anhydride as coupling agent on quality of formed composites

Materials and Methods

Materials used in the study include rice husk and natural bentonite as combined filler, and virgin high density polyethylene (HDPE) as a matrix, xylene 20 % and polyethylene-graft-maleic anhydride (MAPE) with density of 0.92 g/ml, 12 mesh and MW 15,000 as a coupling agent. Rice husk was obtained from paddy field in Aceh Besar region, natural bentonite was taken from Aceh Utara, Aceh Province, while virgin HDPE plastic was purchased commercially. The tools are: stirred reactor consisting of a three-neck flask (Pyrex), motor with stirrer (Fisher Scientific, maximum speed of 250 rpm), oil bath (Corning), Hot press (hand made, temperature range of 29-300 ^{o}C), Crusher and 100-200 mesh size sieve (Macross Testing sieve), Oven with temperature range 25-400 ^{o}C (Gallenkamp), Digital scales, 0-1000 gram (METLER Toledo). Thermometer (0 °C - 200 °C), temperature control (50-500 °C).

Rice husk was initially soaked in hot water at temperature of 100 °C for 2 hours (while mixed). After soaking, the rice husk was dried up to air dry. Later rice husk was grinded and sieved with size of 100-200 mesh and then dried again using the oven at temperature of 105 °C for 24 hours until it reaches the water content of 2% - 3% (Harper and Charles A. 1999). Similarly, natural bentonite was grinded and sieved with size of 100-200 mesh and then dried using the oven at temperature of 105 °C for 24 hours. After that, both rice husk and natural bentonite were kept in a desiccator to prevent contact with the outside air and also to absorb the remaining water vapor contained in rice husk and diatomite before composites manufacturing processes.

Virgin HDPE as much as 40 grams inserted into the three-neck flask and added with 200 ml of xylene as solvent to melt HDPE (Carrol D.R et al. 2001). Furthermore turned on the bath and set the temperature around 145 ^{o}C. Then as much as 60 grams of rice husk/natural bentonite (with rice husk/natural bentonite ratio of 60:40, 70:30, 80:20) was added inside the flask and stirred until homogeneous condition for about 20 minutes with the addition of MAPE coupling agent as much as 2, 3, 4, 5 wt%. Homogeneous mixture was removed from the flask and allowed to cool to evaporate the solvent for 24 hours. Further compression process is carried out by hot press at a temperature of 170 °C for 30 min. Composites then was cooled naturally and later was tested. Tests that conducted in this study were: tensile strength that tested in accordance with ASTM 638-99 Type I; Bending strength using a Universal Testing Machine Electronic System Type: SC-2DE Japan.

Results and Discussion

One of important composite properties that characterized by researchers in their studies about composites is mechanical properties. Mechanical properties of composites are properties that associated with ability of composite to resist external forces that acting on the composite. Mechanical properties of composites that were measured in current research are tensile strength and bending strength. Tensile strength and bending strength of composites that were measured in this study then compared to the standard of SNI 03-2105-1996 and JIS A5908 to understand the quality of produced composites. The results of tensile strength and bending strength of composites on differents variable of composites formation were shown in Figure 1 and Figure 3 below.

Figure 1 showed the effect of addition MAPE on tensile strength at three ratio of rice husk/natural bentonite. The rice husk/natural bentonite ratio was from the highest contain of rice husk to natural bentonite (90:10) until the lowest contain of rice husk to natural bentonite (70:30). The results showed that tensile strength value of all produced composites increased significantly with increase of MAPE percentage at differents rice husk/natural bentonite ratio. The higher tensile strength values were obtained when composites was formed at the lowest rice husk/natural bentonite ratio of 70:30. From the above figure it can be said that rice husk/natural bentonite ratio has a significant effect to improve tensile strength values of formed composites. The increasing amount of natural bentonite in filler of rice husk increased significantly the tensile strength value, for example when rice husk/natural bentonite ratio of 90:10 and addition of 5 wt% of MAPE, the tensile strength value reached only 13.15 MPa and increased sharply to be 16.25 MPa at rice husk/natural bentonite ratio of 80:20 and reached the highest value of tensile strength of 18.20 MPa at rice husk/natural bentonite ratio of 70:30.

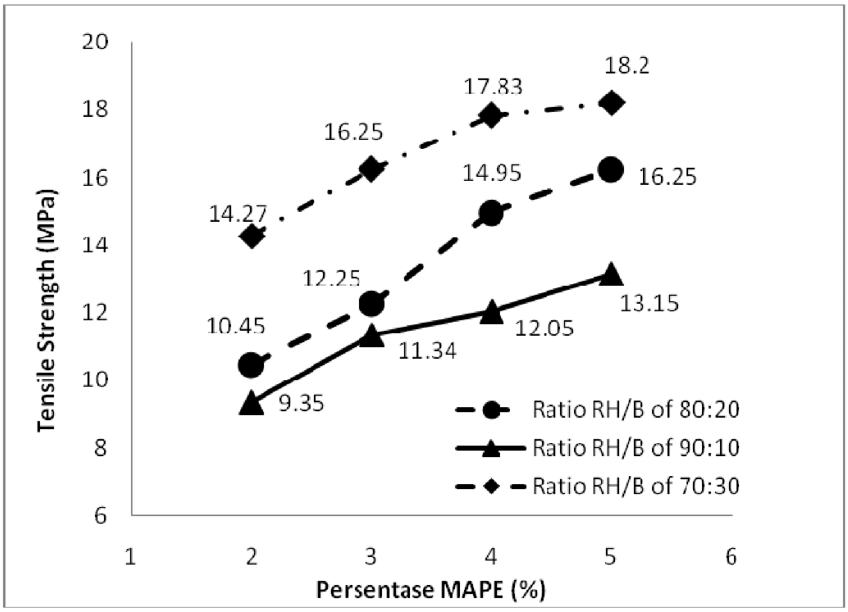

Figure 1. The effect of MAPE percentage and rice husk/natural bentonite ratio on value of tensile strength (RH = Rice husk; B= Natural bentonite)

Some previous studies revealed that MAPE is more effective in improving the bonding between the filler surface and plastic matrix (Mohanty S., Sanjay K. N. 2006). The mechanism of MAPE on the interface in the composite is shown in the following ways.

Cellulosa of filler (-OH group) + coupling agent ⟶ ester lingkage

Above reaction show that reaction occures between the filler containing -OH groups and anhydride groups of coupling agent to form esters lingkage. Moreover, part of HDPE also tends to approach MAPE that has hydrophobic property so that the long chain of HDPE lowering the surface tension. Hydroxyl groups of filler that has hydrophilic properties are not easily mix with the hydrophobic of HDPE matrix. Therefore, the reduce of surface hydrophilicity of filler by coating of filler surface with coupling agent would make easier filler and matrix to combine each other. Furthermore, part of the nonpolar HDPE that combined with MAPE become compatible and also at the same time would reduce the surface energy of the filler, thus increasing the wetness and dispersion in the matrix completely. Mohanty, S. et al. (2007) also found that preparation of composite with MAPE coupling agent can improve mechanical properties compared to preparation of composite without addition of coupling agent. The increase of mechanical properties was due to the increased surface adhesion between filler and matrix with the addition of MAPE.

The above results indicate that coupling agent of polyethylene-graft-maleic anhydride is an effective and suitable coupling agent to produce composites based on a matrix of HDPE polymer. MAPE has been used as a coupling agent to improve the adhesion bonding between ligno-cellulosic fiber derived from biomass-based filler and plastics based matrix (Sameni, J.K. et al. 2004).

In order to understand the effect of addition of natural bentonite into main filler of rice husk, the following figure was created based on previous results that using only rice husk and rice husk/diatomite as filler.

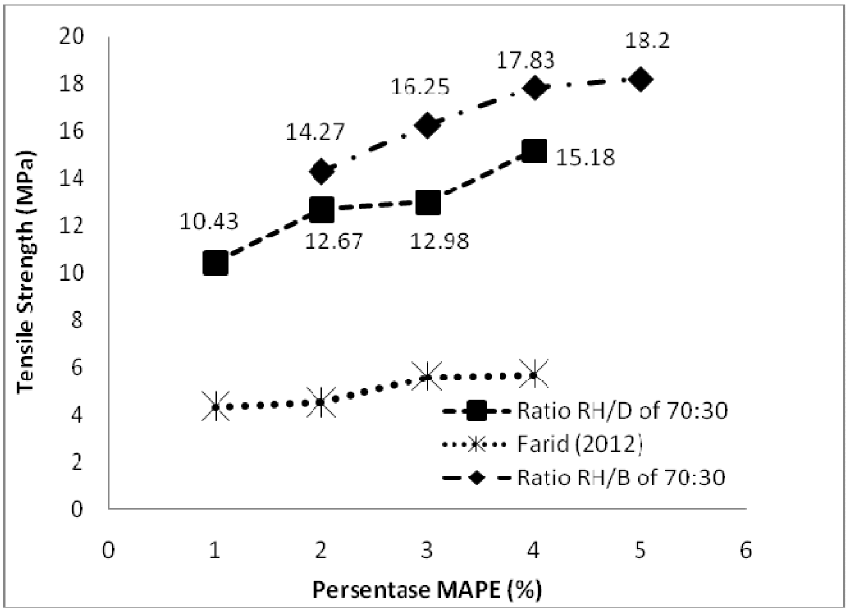

Figure 2. The effect of MAPE percentage and type of used filler on bending strength value (RH = Rice husk; B= Natural bentonite)

Figure 2 showed that composite prepared from filler of rice husk/natural bentonite has the highest tensile strength value among composite prepared from filler of rice husk/diatomite and rice husk itself for similar condition of variables include the used MAPE percentage and ratio of filler. The highest tensile strength value of 17.83 MPa was much higher compared to composite prepared from using rice husk/diatomite as filler with value of 15.18 MPa as found in previous study (Farid M. 2013) and composite prepared from using rice husk as filler with value of 5.69 MPa (Farid M. 2012). The increasing of tensile strength value by addition of 30 wt % natural bentonite in this study reached about more three times compared to previous study that using only rice husk as filler. The high increasing of tensile strength value was due to the effect of natural bentonite addition. The addition of bentonite has increased surface adhesion between filler and matrix and at the same time the pore that still created in composites of previous study has been filled by natural bentonite particles so that the pore become more dense and as result the surface adhesion become stronger. An increase of tensile strength due to the formation of ester bonds between carbonyl groups of the anhydride of coupling agent and the hydroxyl groups of the filler was found by Kamal, B.A, et al. (2008).

Figure 2 also showed that composite prepared from combined filler of rice husk/natural bentonite has higher tensile strength value compared to composite prepared from filler of rice husk/diatomite. It can be seen from from above results that rice husk/natural bentonite formed composite has tensile strength value of 17.83 MPa and rice husk/diatomite formed composite has tensile strength value of 15.8 MPa at the same condition of study. The dominant result of tensile strength value from composite prepared from combined filler of rice husk/natural bentonite might be due to the more hydroxyl groups of the filler created when addition of natural bentonite compared to diatomite. From the above results, it is known that tensile strength value of all prepared composites meets the standards according to SNI 03-2105-1996 where the value of the required minimum tensile strength is 0.15 MPa - 0.29 Mpa (SNI, 2006).

Figure 3 showed the effect of MAPE addition on bending strength at three values of rice husk/natural bentonite ratio. Natural bentonite ratio was added to main filler of rice husk from the highest contain of rice husk (90%) until the lowest contain of rice husk (70%). Similarly to tensile strength value, bending strength value of all produced composites also increased with increase of added MAPE percentage at differents rice husk/natural bentonite ratio. The highest of bending strength value for all prepared composites was obtained at addition of 5 wt% MAPE. The presence of MAPE in the mixture caused the increasing of adhesion bonding between rice husk/natural bentonite with HDPE due to the esterification process between the anhydride groups of MAPE with the hydroxyl group of rice husk. Kamal, B.A, et al. (2008) also found that the addition of coupling agent increase bending strength and composites stiffness significantly. Figure 3 also showed that ratio of rice husk/natural bentonite affected the bending strength value of prepared composite. The bending strength value was improved with increasing the amount of natural bentonite added during composite preparation. The highest bending strength value was obtained at ratio of rice husk/natural bentonite of 70:30 with value of 9.2 MPa at addition of 5 wt% MAPE. This value was much higher compared to previous study without addition of natural bentonite as shown in Figure 4 with value was only 0.35 MPa (Farid M. 2012). However bending strength value of composite prepared from filler of rice husk/diatomite in previous study (Farid M. 2013) was lower than that of composite prepared from filler of rice husk/natural bentonite.

From the above results, it was found that bending strength of composites prepared from filler of rice husk/natural bentonite meet the standard of JIS A5908 but not meet standard of SNI 03-2105-1996. Based on JIS A5908 and SNI 03-2105-1996, minimum standard of bending strength was 8.04 MPa and 9.81 Mpa, respectively.

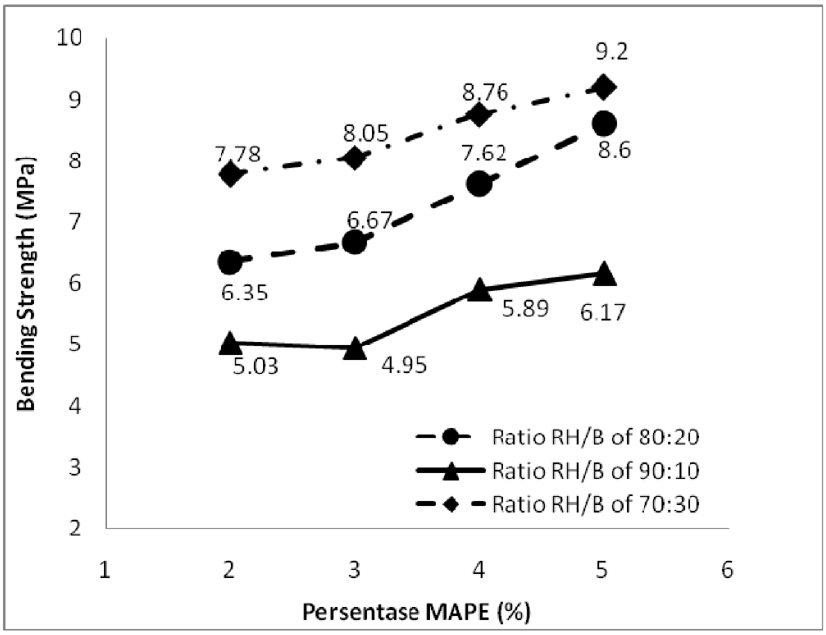

Figure 3. The effect of MAPE percentage and rice husk/natural bentonite ratio on value of bending strength (RH = Rice husk; B= Natural bentonite)

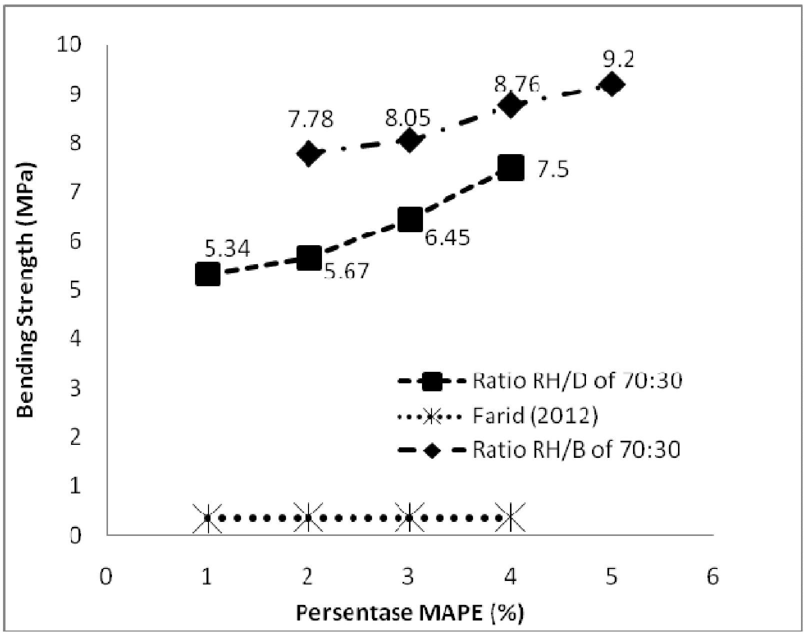

Figure 4. The effect of MAPE percentage and type of used filler on value of bending strength (RH = Rice husk; B= Natural bentonite)

Conclusions

Composites produced from combined filler of rice husk/natural bentonite and matrix of virgin HDPE give much better mechanical properties compared to composites that were prepared by filler of rice husk only and by combined filler of rice husk/diatomite. The highest tensile strength value of 18.2 MPa and bending strength value of 9.2 MPa was obtained on composite prepared by addition of 5% wt of MAPE and at rice husk/natural bentonite ratio of 70:30. Tensile strength value of composites prepared by matrix of virgin HDPE and mixed filler of rice husk/natural bentonite meets the minimum standards of SNI 03-2105-1996 and JIS A5908 but for bending strength value of composite only meet the minimum standards of JIS A5908. The presence of coupling agent and addition of natural bentonite in composites preparation caused the increase of adhesion bond between rice husk and HDPE thereby increasing the mechanical properties of composites.

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