The study on composites formation from HDPE and sawdust/rice husk as raw materials

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Abstract. In this study, composites were prepared using compression molding method from cheap raw materials of high-density polyethylene (HDPE) as matrix and mixture of rice husk and sawdust as filler. The small amount of coupling agent was added during preparation of composites and for comparison composite without addition of coupling agent was also prepared. The coupling agent used in this study was maleic anhydride (MA). The aim of this study is to form composites with and without addition of coupling agent and to know the effect of coupling agent addition and different ratio of matrix and filler toward mechanical properties of produced composites. Composites that contained different ratio of matrix and filler were pressed and heated at temperature of 170°C. The composites were tested its mechanical properties (bending strength and tensile strength) and water absorption capacity. The results indicated that the bending strength of the composite increased with addition of 4 wt% of maleic anhydride and with increased the percentage of filler until 50%. As for the tensile strength, the addition of 4 wt% of maleic anhydride as coupling agent increased the tensile strength significantly. The highest bending strength value of 12.8 MPa was obtained from composite prepared from raw materials contained same amount of matrix and filler and addition of 4 wt% of maleic anhydride. While the highest tensile strength value of 6.1 MPa was obtained from composite prepared from raw materials contained ratio percentage of matrix and filler (40:60 of wt%) and addition of 4 wt% of maleic anhydride.

1. Introduction

Composite board or also known as wood-plastics composite are a product which is widely used since last 40 years [1]. Due to its wide usage, it is constantly changing in term of usage and application. It can be said that composite board has been used as a replacement for regular wood or board. They are used in wide variation, whether used for structural or non-structural application. It can be used indoor as well as outdoor, for example it can be used as construction materials, garden and yard components. It even is used inside the automotive (engine and interior), household item and consumers good. However, the most popular usage of composite are the construction industry and automotive application [1].

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Composite board usually made up of filler which come from wood fiber or organic material and matrix which also known as thermoset or thermoplastic [2]. Thermoset is basically a type of plastic that once cured, cannot be melted by heating such as epoxies and phenolic. While, thermoplastic is basically a type of plastic that can be repeatedly melted. For the matrix, thermoplastics are more preferable due to its ability to be melted multiple times allowing it to be shaped and recycled. The composite have characteristics such as low density, low manufacturing cost, renewable and recyclable [3].

Composite board can be derived from used item. For example, the organic material can be come from used wood particle or any plant waste, such as the sawdust, rice husk and many more. The matrix is not necessarily comes from virgin thermoplastics. Used or recycled thermoplastics can also be used as the matrix [4]. These properties allow the composite to become more popular. Now, waste from organic materials can be easily found. Sawdust is usually left behind at the sawmill. The rice husk and rice stalk from the rice field usually being burned and causing air pollutions. With formation of composites with these used materials, waste from those industries can be minimized, and pollution can be reduced. Those biomass materials of sawdust and rice husk have several advantages such as low density, low cost, renewable and recyclable and biodegradability properties [5, 6].

Despite its big popularity, the composite board is usually not strong enough compared to regular board, due to its low adhesion between the matrix and the filler. Most of the matrix, especially thermoplastics, is non-polar (hydrophobic) substances, which are not compatible with polar (hydrophilic) wood fibers and, therefore, resulting the poor adhesion between polymer and wood fiber. In order to improve the adhesion between the wood fibers and polymer, addition of coupling agent is needed [6]. Coupling agent is basically a chemical that is added to the mixture in small quantities, so it allow bonding to occur between its surface with the other surface such as the polymer and the wood fibers. The coupling agent will encourage the spreading of particle that will increase the adhesion between the polymer and wood fibers. So, the addition of coupling agent will increase the strength of the composites that will allow the composite to be used even more frequently in many applications.

There are several studies have been conducted by researchers regarding the formation of composite by using the sawdust and rice husk separately [4, 5, 6]. But there is no research has been done for the formation of composite by using mixture of sawdust and rice husk. So, due to that, mixture of sawdust and rice husk is selected as the fillers. As for coupling agent, the most popular coupling agent of maleic anhydride (MA) is selected. 4 wt% of maleic anhydride was added into mixture of matrix and filler because according to [6, 7], the most optimum value in term of mechanical properties was obtained when coupling agent was added at 4 wt% of the matrix and fillers weight. Different ratio of matrix and filler was performed, because according to [8], similar ratio of matrix and filler was the most suitable for the melt-blending formation.

The aim of this study is to form composites from high-density polyethylene as matrix and mixture of rice husk and sawdust as the filler with and without addition of maleic anhydride as coupling agent and to know the effect of addition of coupling agent and different ratio of matrix and filler toward mechanical properties and water absorption capacity (%) of produced composites.

2. Methodology

2.1. Equipments and Materials

The equipment used for this study included stirred reactor consisting of a beaker (Pyrex), motor with stirrer (Fisher Scientific, max. speed of 250 rpm), oil bath (silicone oil), hot press (Boy22A), grinder and +80-200 mesh size of sieve with shaker, oven, digital scale, and thermometer (0-300°C). The materials used in this study were rice husk and sawdust as the filler, high density polyethylene (Sigma-Aldrich) as matrix, xylene as solvent and maleic anhydride (Sigma-Aldrich) as the coupling agent. Rice husk was obtained from BERNAS Sekinchan and sawdust was obtained from a workshop in Faculty of Civil Engineering, UiTM Shah Alam.
2.2. Formation of Composites
2.2.1. Preparation of Matrix and Filler
In order to eliminate the extractive substance that may still be contained inside the filler of rice husk and sawdust, it was soaked with hot water at temperature of 100°C for about 2 hours. During soaking, the filler was stirred at the medium speed. After that, the filler was dried up in the oven for 24 hours. Later, the filler was grinded and sieved with +80-200 mesh size sieves [7]. Then, the filler was dried again by using oven at temperature of 110°C for 24 hours in order to remove the excess moisture content to be less than 8% by weight [9]. After dried for 24 hours, the filler was stored in the desiccator to prevent any moisture to absorb into the filler.

2.2.2. Formation of Composites
Composites were formed by using compression molding method. Initially, 35 g of virgin HDPE was placed inside the flask, and 100 mL of xylene was added into the beaker slowly. The flask was immersed into the oil bath that was heated at the temperature of 145°C. After the matrix was fully melted, 15 g of filler comprised of 7.5 g of dried sawdust and 7.5 g of dried rice husk, was added to the flask and stirred using stirrer until both matrix and filler were fully homogenized for about 20 minutes. Maleic anhydride with amount of 4% by weight was added to the mixture inside the flask. The ratio of matrix and filler were varied at mass ratio of 70:30, 60:40, 50:50 and 40:60. After the mixture was fully homogenized, the mixture was removed from the flask and allowed to cool down naturally until room temperature and until all the solvent evaporated. For further compression, it was compressed by using hotpress at temperature of 170°C for 30 minutes. The composites then cooled naturally and tested. Another composite with the same variation of ratio of matrix and filler was also prepared but without addition of maleic anhydride for comparison.

2.2.3. Testing and Analysis of Prepared Composites
Mechanical properties of composites that include tensile and bending strength were tested using Universal Testing Machine Model SHIMADZU in Concrete Lab, Faculty of Civil Engineering, UiTM Shah Alam. Water absorption capacity test was performed as procedure written in SNI 03-2105-2006 [10]. The results of bending strength, tensile strength and water absorption capacity value from this study were later compared to the standard of SNI 03-2105-2006 (the quality of particle board). According to SNI 03-2105-2006, the minimum standard of bending strength and the tensile strength is 9.81 MPa and 0.15 MPa, respectively and the water absorption capacity value was less than 12 %.

3. Results and Discussion
3.1. Mechanical Properties of Prepared Composites
Maleic anhydride is a bridge that connecting the polar of wood fiber with the nonpolar of polymer which are not compatible with polar (hydrophilic) wood fibers and, therefore, resulting the better adhesion between polymer and wood fiber. Theoretically, mechanical properties of composites are associated with ability of composite to resist external forces that acting on the composite.

Figure 1 shows results of tensile strength for all composites with and without addition of maleic anhydride at different ratio of matrix of HDPE and filler of rice husk and sawdust. According to figure 1, the increasing of percentage of filler in the mixture of matrix and filler resulted in increasing of tensile strength of the prepared composites. The highest tensile strength of 6.1 MPa was obtained when the percentage of filler was at 60% with the addition of maleic anhydride. The lowest tensile strength of 1.3 MPa was obtained on composite contained 30 wt% filler without addition of the maleic anhydride.

From the above results, it indicated that tensile strength of all composites by using HDPE met the standard of SNI 03-2105-2006. The highest tensile strength of 6.1 MPa in this study was a bit higher compared to similar research using raw material of only rice husk as filler with tensile strength of 5.75 MPa in our previous paper [7] and with tensile strenght of 5.69 MPa in [6]. The highest tensile
strength of composite in both previous papers was also found on samples after addition of 4 wt% of coupling agent.

![Graph showing effect of percentage of filler on tensile strength](image)

**Figure 1.** The effect of percentage of filler (%) toward tensile strength value of prepared composites with addition and without addition of maleic anhydride (MA) coupling agent.

Figure 1 also showed that the prepared composite with addition of 4 wt % of maleic anhydride was stronger than the prepared composite without addition of maleic anhydride. The presence of small amount of maleic anhydride induced the stickiness between rice husk/sawdust that has properties as hydrophilic and HDPE as hydrophobic material was improved. The maleic anhydride acted as bridge link between both matrix and filler to increase adhesion properties inside the composite and finally increased the tensile strength of composite. The similar structure and formation mechanism of this prepared composite were studied previously by another researcher [11]. However, there was slight inconsistency the result with 40% percentage of filler, where the tensile strength was higher at sample without addition of maleic anhydride. This inconsistency may occur due to the presence of air bubble inside the prepared composite. Slight inconsistency in the thickness of the composite may also affect the results. This inconsistency of thickness may due to the method of compression molding; where it does not really control the thickness compared to the injection molding method [1, 12].

Figure 2 showed results of bending strength for all composites with and without addition of maleic anhydride at different ratio of matrix of HDPE and combined filler of rice husk and sawdust. Figure 2 showed that the presence of maleic anhydride content in the mixture resulted in increasing of bending strength of the prepared composites almost twice. The highest bending strength was obtained when the percentage of filler was 50 wt% with the addition of 4 wt % maleic anhydride and the value was 12.8 MPa. This bending strength value was higher than that of a minimum standard of 9.81 MPa as stated in SNI 03-2105-2006. However, all bending strength value of the prepared composites without addition of maleic anhydride did not meet SNI 03-2105-2006.

As in tensile strength, for the bending strength value there was slight inconsistency in the result. At 40% filler, the bending strength was lower on a sample with addition of maleic anhydride. This inconsistency may occur due to the presence of air bubble inside the prepared composite and also due to the thickness of the composite as explained previously. To understand the resistance of composite when contacting to moisture or wet environment, so all prepared composites were performed the water absorption testing by immersing composites in water. The testing results of water absorption were shown in figure 3. Composite board is a substance that has the hygroscopic properties so that it is able
to absorb moisture from the surrounding environment to achieve equilibrium and likewise when composite board was immersed in water it would expand. Water absorption capacity was determined based on the weight of the specimens after being immersed in water in accordance with the method requirement in SNI 03-2105-2006.

![Figure 2](image-url)

**Figure 2.** The effect of percentage of filler (%) toward bending strength value of prepared composites with addition and without addition of maleic anhydride (MA) coupling agent.

![Figure 3](image-url)

**Figure 3.** The effect of percentage of filler (%) toward water absorption capacity (%) value of prepared composites with addition and without addition of maleic anhydride (MA) coupling agent.
The results showed that the water absorption capacity from the prepared composites with addition of 4 wt% of maleic anhydride on average range from 7.0% - 11.6%, which these results indicated that the water absorption capacity in all prepared composite board with addition of coupling agent met SNI 03-2105-2006. On the other hand, the water absorption capacity of composites without addition of maleic anhydride only met the standard on samples with percentage of filler 30% and 40%. Therefore, it can be seen clearly that the composites with addition of maleic anhydride had a better properties of water absorption resistance compared to the composites without addition of maleic anhydride [13].

4. Conclusion
The bending strength and the tensile strength of the composite increased with addition of maleic anhydride coupling agent and also increased as the percentage of filler increased. The addition of 4 wt% of maleic anhydride coupling agent affected significantly the mechanical properties of prepared composite. The presence of maleic anhydride coupling agent in composites resulted in increasing adhesion properties between rice husk/sawdust and HDPE thereby increasing the mechanical physical properties of composites.

Tensile strength, bending strength and water absorption capacity values of prepared composites with addition of 4 wt% of maleic anhydride met the minimum standards of SNI 03-2105-2006. The highest tensile strength and bending strength of composites with the addition of 4 wt% maleic anhydride was 6.1 MPa and 12.8 MPa, repectively. In addition, the water absorption capacity of composites with addition of 4 wt% of maleic anhydride was in the range of 7.0% – 11.6%.

References
[1] Schwarzkopf M J and Burnard M D 2016 Wood-Plastic Composites—Performance and Environmental Impacts, in Environmental Impacts of Traditional and Innovative Forest-based Bioproducts, Springer 19-43.
[2] Clemons C 2002 Forest Products Journal 52 (6) 10.
[3] Hung K C, Yeh H, Yang T C, Wu T L Xu J W and Wu J H 2017 Polymers 9 726
[4] Mulana F, Hisbullah dan Iskandar 2011 Jurnal Rekayasa Kimia dan Lingkungan 8 (1) 17-22
[5] Moreno D D P and Saron C 2017 Composite Structures 176 1152-1157
[6] Mulana F, 2012 Composites prepared from rice husk and recycled/ virgin HDPE with addition of coupling agent The Proc. of 2nd AIC Unsyiah & 8th IMT-GT Uninet Biosciences Conference, 22-24 November 2012, Banda Aceh, Indonesia
[7] Mulana F, 2014 The effect of MA/MAPE combination as coupling agent in formation of rice husk and recycled HDPE-based composites Proc. of the 4th AIC Unsyiah in conjunction with 9th AIWEST-DR, October 22-24, 2014, Banda Aceh, Indonesia
[8] Lu J Z, Wu Q and McNabb H S 2007 Wood and Fiber Science 32 (1) 88-104.
[9] Schmidt E 1993 Methods for preparation of composite materials US Patent 5,217,655
[10] Badan Standarisasi Indonesia (BSN) 2006 Papan Partikel SNI 03-2105-2006
[11] Yang H S, Kim H J, Park H J, Lee B J and Hwang T S 2007 Composite Structure 77 45 - 55
[12] Yam R C M and Mak D M T 2014 Journal of Cleaner Production 67 277-284
[13] Gilormini P and Verdu J 2018 Polymer 142 164 - 169