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Effect of Corn Husk Fiber Addition on Mechanical Properties of High-Density Polyethylene (HDPE) Composites with Aluminum Oxide (Al2O3) and Maleic Anhydride (MAH) as Coupling Agent

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ABSTRACT

The application of composites is widely used in the automotive field. HDPE, a versatile polymer, is frequently employed as a composite matrix, combining with other materials to produce unique mechanical properties and characteristics. Its widespread application in the automotive sector includes the manufacturing of bumpers and car interiors, where its distinct properties enhance the overall performance and functionality of these components. Corn husk fiber waste is a natural material that has not been utilized properly, so the use of corn husk fiber as reinforcement in composites is an alternative to waste problems in the environment. Aluminum Oxide (Al_2O_3) as a filler with another name alumina, possesses strong tensile strength and thermal stability, and it is anticipated to lower production costs while enhancing composites mechanical qualities. Maleic anhydride (MAH), a coupling agent, is applied to the matrix, reinforcement, and filler to increase their adherence. The manufacture of HDPE matrix composite with the addition of corn husk fiber and MAH is expected to improve the mechanical properties of the composite. The composition of HDPE: corn husk fiber is 90:0, 85:5, and 80:10 (%wt), as well as the composition of 5%wt alumina and 5%wt MAH in each sample. Making composite pellets using a compounder, making composite sheets by hot press method using a manual forming machine. Tensile strength and hardness measurements were taken to evaluate material performance. Tensile strength testing was carried out using a Universal Testing Machine (UTM), crystallinity testing using Differential Scanning Calorimetry (DSC) and testing using Near-Infrared (NIR). The tensile strength test results obtained the highest tensile strength value in the composition of HDPE: corn husk fiber 90: 0%wt of 17.67 MPa. The crystallinity test results obtained the highest degree of crystallinity in the composition of HDPE: corn husk fiber 90: 0%wt of 67.88%. The incorporation of corn husk fibers into the composite material significantly affects its mechanical properties, particularly the tensile strength and crystallinity, with higher fiber content leading to a notable reduction in these properties. SEM testing was also carried out to determine the morphological shape of the composite.

I. Introduction

Along with the development of technology, polymer applications, especially composites, are increasingly widely used, especially in the automotive industry. Composites are materials consisting of two or more constituent materials that have the properties of each material in the end result [1].

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The development of composite materials is one of the important aspects in the material industry to produce stronger, more durable, and environmentally friendly products, so that improvements in the physical and mechanical properties of composites continue to be made in order to obtain characters according to industry needs [2]. Composite materials generally consist of a matrix, polymeric adhesive material and reinforcement in the form of natural fibers [3]. The matrix used in this research is a thermoplastic, namely High-density Polyethylene (HDPE)[4]. High-Density Polyethylene (HDPE) is one of the thermoplastic polymers that has strength and corrosion resistance so that HDPE applications are widely used in the automotive industry [5], but HDPE has the inability to interact with natural fibers so that the addition of coupling agents is needed. Research has focused on enhancing the mechanical properties and durability of HDPE composites by incorporating natural fibers as reinforcement materials. The use of natural fibers in composite materials is an alternative because it is environmentally friendly and recyclable [6].

In this study, corn husk fiber was used as a natural fiber. The incorporation of corn husk waste fibers into composite materials is anticipated to contribute significantly to environmental sustainability by minimizing waste disposal and enhancing the mechanical properties of these composites [7]. The addition of MAH coupling agent in the composite to improve the bond between fiber and matrix [8]. MAH can increase the tensile strength of the composite and the enthalpy value of melting with a percentage of 5%. In addition, the addition of 5% MAH to polyethylene composites shows higher tensile strength values and degree of crystallinity [9]. The addition of additives when making composites in automotive component applications is a current consideration [10]. Al_2O_3 is known as an effective reinforcement in polymer composites. The addition of Al_2O_3 by 5%, 10%, and 15%, showed that the 5% Al_2O_3 composition showed the highest sample melting enthalpy compared to the 10% and 15% Al_2O_3 compositions [11].

Based on the description above, researchers conducted research by making high-density polyethylene (HDPE)/corn husk fiber (SKJ) composites with the addition of alumina powder (Al_2O_3) as filler and maleic anhydride (MA) as coupling agent. The composition variation is the first sample consists of 90 HDPE: 0 SKJ: 5 Al_2O_3: 5 MAH %wt, the second sample 85 HDPE: 5 SKJ: 5 Al_2O_3: 5 MAH %wt, the third sample of 80 HDPE: 10 SKJ: 5 Al_2O_3: 5 MAH %wt. The impact of varying corn husk fiber content in HDPE composites on their tensile strength and crystallinity was investigated to assess their potential for use in automotive components. Tests using NIR to determine the chemical interactions between the composite constituent materials. SEM testing was carried out to determine the morphological shape of the composite so that the analysis of fiber distribution in HDPE matrix is known.

II. Method

A. Delignification Process of Corn Husk Fiber

The delignification process uses a 10%wt KOH solution. Starting with weighing KOH as much as 100 grams, then KOH that has been weighed is dissolved with 1000 ml of distilled water. The corn husk fiber, which had been separated, was placed in a beaker with a KOH solution. It was then soaked for 4 hours, with stirring every hour, and the pH was measured using universal pH meter paper.

B. Process of Plastic Compounding

The plastic compound was made using HDPE polymer, corn husk powder, aluminum oxide powder, and maleic anhydride. The plastic compound is made using a compounder machine according to the first sample variation consisting of 90 HDPE: 0 SKJ: 5 Al_2O_3: 5 MAH %wt, the second sample of 85 HDPE: 5 SKJ: 5 Al_2O_3: 5 MAH % wt, the third sample 80 HDPE: 10 SKJ: 5 Al_2O_3: 5 MAH %wt. The materials that have been prepared are weighed according to the ratio. The ingredients will be combined in a beaker and agitated with a stirring rod. The blended ingredients will be introduced into the compounder. The output from the strand pelletizer will be gathered in a beaker until it stops flowing, which can be identified by the distinct sound of the extrudate breaking as it comes out of the die.

C. Composite Preparation

The plastic compound material was used to create the composite sheet using a compounder. After pouring the extrudate pellets into a metal plate mold, they were flattened to cover the entire side of the metal plate. The hot press method is employed to produce composite sheets, where the material is pressed using a manual forming machine.

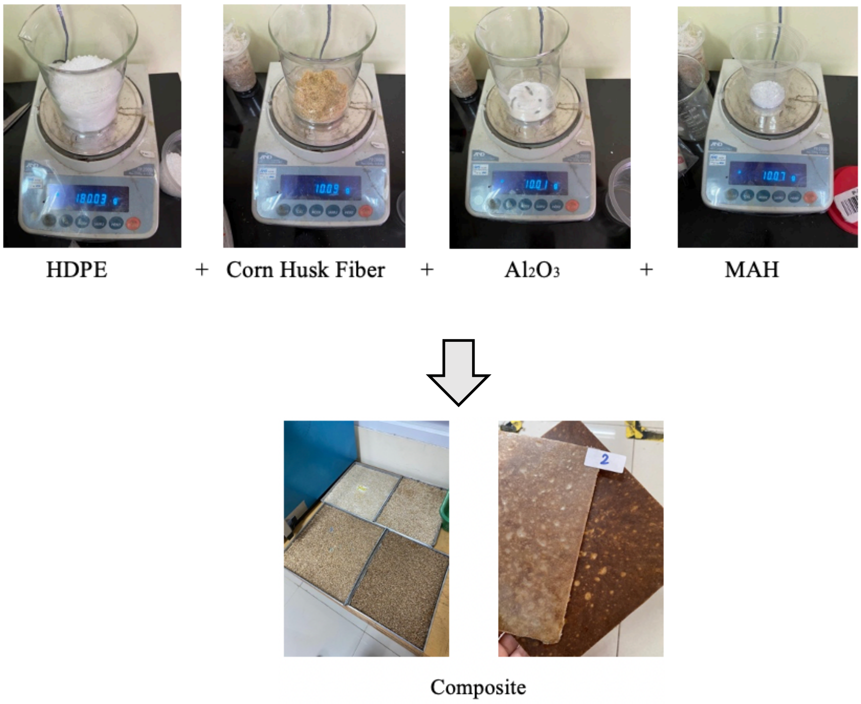

Fig.1 The process of produce composite sheets

D. Composite Characterisation

Composites that have been made into test specimens will be used for the testing process. The tests carried out for the composite are tensile strength test with Universal Testing Machine (UTM) at Polymer Laboratory of STMI Polytechnic Jakarta and crystallinity test with Differential Scanning Calorimetry (DSC) at Instrumentation Laboratory of STMI Polytechnic Jakarta. NIR testing was conducted at the University of Indonesia. Morphological test using SEM was conducted at BRIN.

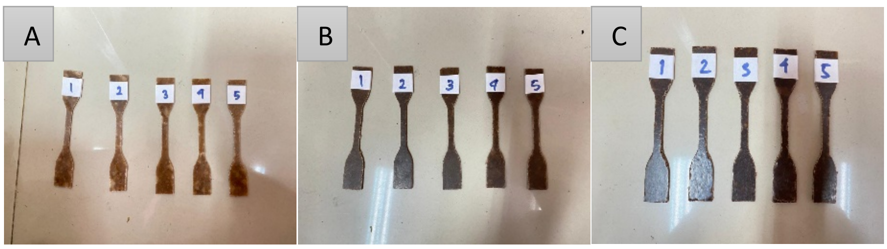

Fig 2. Specimens before tensile strength test (A) first sample variation consisting of 90 HDPE: 0 SKJ: 5 Al_2O_3: 5 MAH %wt; (B) second sample of 85 HDPE: 5 SKJ: 5 Al_2O_3: 5 MAH % wt; (C) third sample 80 HDPE: 10 SKJ: 5 Al_2O_3: 5 MAH %wt

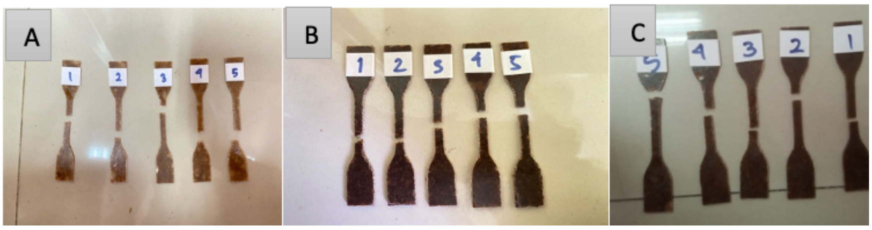

Figure 3. Specimens after tensile strength test (A) first sample variation consisting of 90 HDPE: 0 SKJ: 5 Al_2O_3: 5 MAH %wt; (B) second sample of 85 HDPE: 5 SKJ: 5 Al_2O_3: 5 MAH % wt; (C) third sample 80 HDPE: 10 SKJ: 5 Al_2O_3: 5 MAH %wt

III. Results and Discussion

A. Results of Tensile Strength Testing

Tensile strength testing has an important role in revealing the extent to which the addition of corn husk fiber, Al_2O_3, and MAH affects the tensile strength of HDPE polymer composites. Table 1 below shows the data obtained when testing the tensile strength with variations of 0%wt corn husk fiber, 5%wt, and 10%wt composites with the addition of 5%wt Al_2O_3 and 5%wt maleic anhydride per sample. The results of the tensile strength testing of composites with 0%wt corn husk fiber composition variation were 17.67 MPa; 5%wt composition was 7.13 MPa; and 10%wt composition was 5.67 MPa. Based on the tensile strength value, it can be seen that the 0%wt corn husk fiber composition variation gives the highest tensile strength value of 17.67 MPa.

Table 1. Variation of corn husk fiber on tensile strength of HDPE composites

Sample HDPE:SKJ:Al_2O_3:MAH	**Average Tensile Strength (Mpa)**
90:5:5:0 %wt	17,67
85:5:5:5 %wts	7,13
80:10:5:5 %wt	5,67

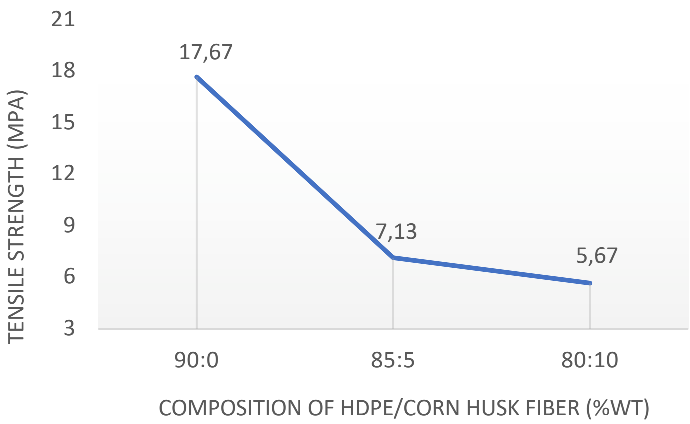

Fig 4. Diagram of tensile strength values against sample composition variation

Figure 1 shows the tensile strength value of the composite with 0%wt corn husk fiber composition variation giving the highest tensile strength value of 17.67 MPa. The decreased tensile strength can also be caused by the lack of homogeneity of the resulting composite, the fibers are not evenly distributed resulting in agglomeration. Agglomeration of corn husk fibers results in a weak adhesion force between the fibers and the thermoplastic matrix [12]. Other research also shows that the higher the fiber addition, the lower the homogeneity of the composite [13] [14]. This can also be due to the cohesion force between the constituent components of the composite is greater than the adhesion force of the constituent components. The possibility that corn husk fiber as reinforcement has an uneven contribution during compounding and when making test specimens there are voids or air bubbles. Voids affect the quality of a composite, the more voids indicate the quality of the composite is decreasing, because voids can be a concentration of stress when the composite receives a force [10]. Initially, the addition of MAH can improve the adhesion between corn husk fiber and polyethylene matrix due to better solubility and reactivity. However, if the MAH concentration is too high, it may cause negative effects such as:

1. Over-crosslinking: The presence of an overly high MAH concentration may result in an excessive crosslinking of polyethylene chains. [15]. The ductility of the material may be diminished, leading to increased brittleness.
2. Decreased Molecular Mobility: Excessive adhesion between the fibers and the matrix can inhibit the mobility of molecules in the polyethylene matrix [16]. This can result in a decrease in elasticity and tensile strength.
3. Increased Interface Layer Thickness: An increased MAH concentration may cause the interface layer between the matrix and the fibers to become thicker. [17]. Tensile strength values could be lowered as a result of this interfering with the force transfer between the fibers and the matrix. [18].

Therefore, while coupling agents like MAH are required to improve the contact between the fiber and matrix, it is important to find the right concentration. Due of the previously listed negative impacts, raising the MAH concentration may eventually no longer have a positive improvement and may instead cause the tensile strength value to decrease. Consequently, in order to find the MAH concentration that best balances the mechanical and adhesive capabilities of polyethylene composites, experimental research is required.

B. Stability Testing Results

The crystallinity test results of polymer composites have great relevance in understanding the structure and mechanical properties of the material [19]. Crystallinity refers to the degree of crystallinity in a polymer material, which can affect the mechanical, thermal, and optical properties of the composite. Table 2 shows the data from the thermal properties testing using DSC to determine the degree of crystallinity of the enthalpy value of HDPE/corn husk fiber composite with variations of 0%wt, 5%wt, and 10%wt corn husk fiber and the addition of 5%wt alumina and 5%wt maleic anhydride per sample.

On the second heating of the thermogram, the enthalpy of melting value of the sample (ΔHm) was detected. The enthalpy of melting value of the composite crystallinity testing with 0%wt corn husk fiber composition variation was found to be 198.9 J/g; 5%wt composition was 182.2 J/g; 10%wt composition was 173.6 J/g. Based on the value of the enthalpy of melting (ΔHm), the highest value was obtained in the 0%wt corn husk fiber composition variation, which was 198.9 J/g.

Table 2. Variation of composite composition on crystallinity value.

Sample HDPE:SKJ:Al_2O_3:MA	**ΔHm (J/g)**	**Xc (%)**
90:0:5:5 %wt	198,9	67,88
85:5:5:5 %wt	182,2	62,18
80:10:5:5 %wt	173,6	59,25

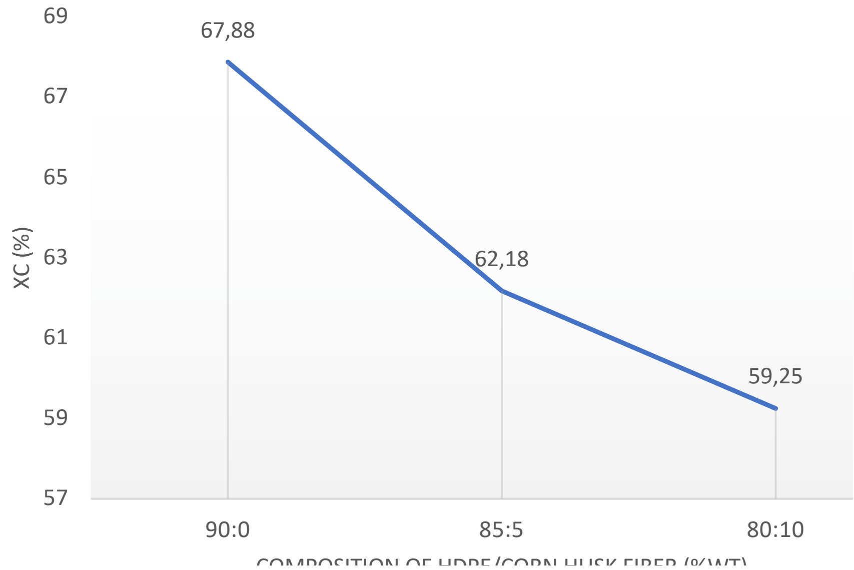

Fig 5. Diagram Variation of composite composition on crystallinity value

The degree of crystallinity indicates the number of crystals formed in the composite [20]. The degree of crystallinity has a correlation with mechanical properties, one of which is tensile strength [21]. The higher the degree of crystallinity of a composite, the higher its tensile strength [22]. The crystallinity test results can help identify the effect of the addition of corn husk fiber, High-Density Polyethylene (HDPE), Aluminum Oxide (Al_2O_3), and Maleic Anhydride (MAH) on the crystal structure of the composite. Different material compositions can result in variations in the degree of crystallinity, which affects the mechanical and thermal properties of the composite. In this study, the highest crystallinity was found in the 0%wt corn husk fiber composition, because corn husk fiber can inhibit the crystal formation process in the composite [23].

This is in line with research [24], that the value of the degree of crystallinity of 5%wt corn husk fiber is 90% and 10%wt is 83%, indicating that the more weight % of corn husk fiber in the composite can reduce the degree of crystallinity. Another study conducted by [25] that the decrease in the degree of crystallinity also occurs that the greater the weight % composition of corn stalk powder in LDPE composites which reduces the value of the degree of crystallinity.

C. Functional Group Testing Result

Functional group testing uses NIR to find out information about the chemical composition, molecular structure, and interactions between components in polymer composites. The results of NIR analysis can give an idea of how well these components interact and are homogeneously mixed in the polymer matrix. An even composition can result in superior mechanical properties. These interactions can influence the quality of the interparticle bond, affecting the strength of the composite structure and optimising its mechanical properties.

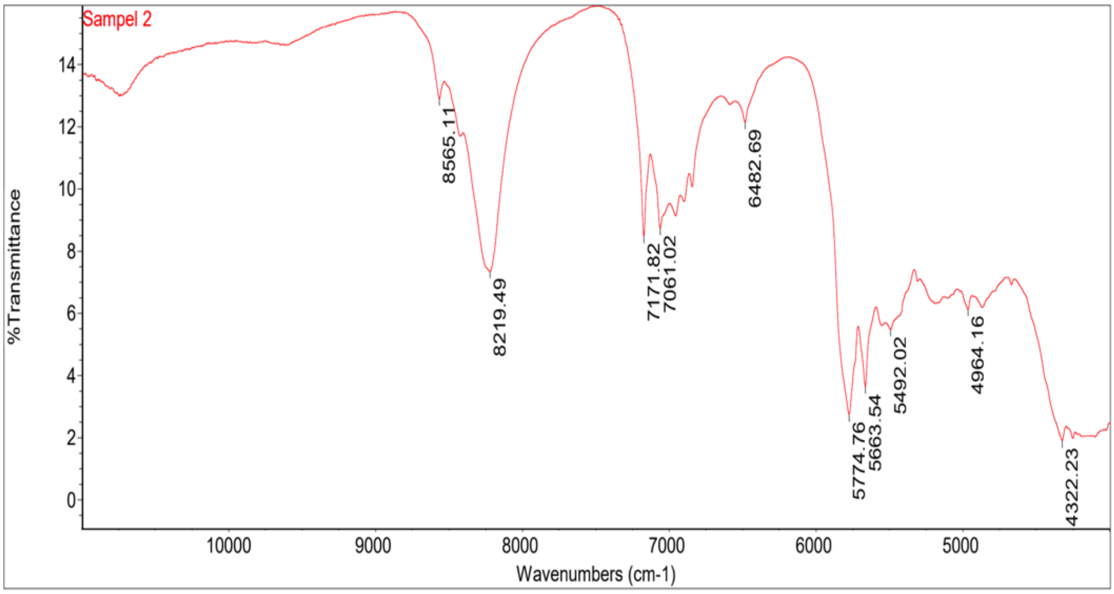

Table 3. NIR functional groups of HDPE/SKJ/Al_2O_3/MAH Composite

Wavelength	Intensity	Functional Groups
4322.23	1.884	$-CH_2-$/-CH-/-C=C-
4964.16	6.102	-NH- / -OH- [26]
5492.02	5.468	-OH-
5663.54	3.613	-CH- dan $-CH_2-$
5774.76	2.735	-CH- dan $-CH_2-$
6482.69	12.139	-OH-/-C-O-
7061.02	8.714	-CH-
7171.82	8.436	-CH-
8219.49	7.335	-CH-
8565.11	12.882	-CH-

Based on the data in Table 3, there are several functional groups in the composite, including: -CH_2=CH_2- at a wavelength of 4322.23 cm^{-1} [26], which indicates the presence of HDPE. At 5212.31 cm^{-1} shows -OH-, at 5663.54 and 5774.76 cm^{-1} shows -CH- and $-CH_2-$ groups [27] are functional groups present in corn husk fiber. The -C-O- group also shows the functional group in MAH. The above functional groups indicate that there are interactions between the components that make up the composite. This information can help researchers and industry in designing and optimizing polymer composite formulations that meet the desired mechanical and performance requirements.

D. Morphological Testing Results

SEM tests were conducted to determine the distribution of corn husk fibers and Al_2O_3 particles in the HDPE matrix. An even distribution of fibers and filler particles that are well bound to the matrix can produce a homogeneous composite. The homogeneous distribution also favours the improvement of the mechanical properties of the composite, as each part of the material faces the load in a uniform way.

SEM analysis allows evaluation of the adhesion between corn husk fibers, HDPE matrix, and Al_2O_3 particles. Good adhesion between fibers, matrix, and filler is key to achieving high tensile strength and stiffness in composites. The visible fiber surface is firmly bonded to the matrix and filler demonstrating the effectiveness of the addition of MAH as a coupling agent. Figure 4 shows the morphological results of the HDPE/SKJ/Al_2O_3/MAH composite with 10000x magnification.

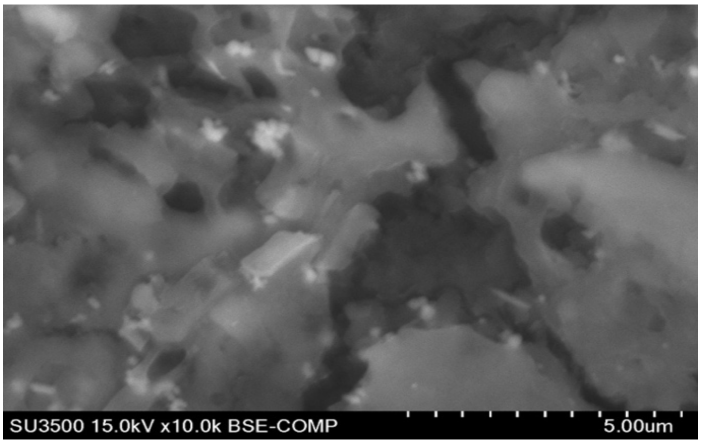

Fig 7. Morphology of HDPE/SKJ/Al_2O_3/MAH composite with 10000x magnification

Testing the surface condition (morphology) of HDPE/SKJ/Al_2O_3/MAH composites with 10000x magnification was carried out to determine the homogeneity of the distribution of coffee grounds waste as filler and the role of MAH as a coupling agent. The morphological test results show that there are several voids in the composite which show that the composite constituent components are less homogeneous, this can be caused during the mixing process. This inhomogeneous mixing is one of the causes of decreased mechanical properties with the addition of corn fiber, Al_2O_3, and MAH.

SEM (Scanning Electron Microscopy) test is a microscopy technique used to explore the surface structure of samples with high resolution. In the context of research into the distribution of corn husk fibers and Al2O3 particles in a polyethylene (HDPE) matrix, SEM tests are highly relevant to provide an in-depth visual understanding. SEM tests provide an in-depth and detailed view of the distribution of fibers and particles in polyethylene composites, which can be the basis for the development of more mechanically and structurally superior materials.

IV. Conclusion

Based on the research results and analysis of the test data, it is concluded that the addition of Maleic Anhydride (MAH) to composite that contains polyethylene: corn husk fiber: Al_2O_3: MAH decreases the tensile strength value. The best composition for tensile strength value with the addition of MA 0% weight obtained the highest result of 13.47 MPa. The effect of Maleic Anhydride (MAH) addition on polyethylene: corn husk fiber: Al_2O_3: MAH composite decreases the degree of crystallinity. The best composition for the degree of crystallinity value with the addition of MAH 0% weight obtained the highest result of 42.16%.

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