Characterization of low-density polyethylene (LDPE)/carbon black (CB) nanocomposite-based packaging material

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Abstract. Carbon black (CB) has the ability to be used as a filler to improve the thermal and mechanical properties of the packaging material. This study aimed to determine the effect of CB composition variations as a filler on low-density polyethylene (LDPE) nanocomposite to physical properties, mechanical properties, and thermal properties. The fabrication of LDPE/CB nanocomposite-based packaging material was performed by hot press and cold press method with LDPE/CB composition variations of 95: 5, 90:10, 85:15, and 80:20 \text{ wt.} The effects of CB as filler on LDPE thermoplastic were characterized using FTIR, XRD, SEM, UTM, and TGA. The result of IR spectra analysis showed that the interaction between LDPE and CB in the solution was limited to physical bonds, the analysis using XRD generally follows the LDPE diffraction pattern. The most optimum composition result obtained from the variation of 5 wt CB, where the surface morphology using SEM of it tends to agglomerate, the mechanical analysis using UTM showed a tensile strength of 21.45 MPa, elongation at 436.21 mm, and modulus young 246.22 MPa, and thermal analysis using TGA showed changes mass at a decomposition temperature of 488.70 °C.

I. Introduction
In recent years the need for polymers in human life has increased rapidly, even including as a primary need, one of which is shown by its use as packaging (1). Packaging is a much-needed component of food manufacturing and food supply processes. Packaging serves to control product exposure to the effects of oxygen, light, steam, and water and bacterial contamination. The packaging sector is one of the most important global industries representing about 2% of Gross National Product (GNP) from developed countries (2). This is shown in a brief data evaluation in which developed countries show nearly 98% of packaging products, especially in widely used food packaging consisting of plastic, paper, glass, metals, and polymer matrix composites (3).

Polymer matrix composite (PMC) is the most promising material in this century, the structure consists of two phases, namely filler (reinforcement) in the form of carbon particles and the matrix in the form of polymers (4). PMC is one of the alternative materials used in packaging materials because it has a lightweight, easy to form, high strength and toughness, is not easy to absorb moisture and high corrosion.
resistance. In general, packaging material has several disadvantages, namely having a low melting point and tensile strength and easily deforming which causes early damage to the packaging material (5).

Therefore, good quality packaging materials are needed which have high melting point properties to be resistant to temperature and have good mechanical properties. A way to improve the two properties of the packaging material is by adding Carbon Black (CB) to the Low-Density Polyethylene (LDPE) material. CB is a material consisting of 97% carbon elements and (0.2 - 1.5%) elements of oxygen and hydrogen. CB has a very small particle diameter and the inter-particle arrangement is mutually tight to each other so that there is cohesiveness bond between CB and LDPE matrix which forms a strong covalent bond (6). This is consistent with several studies which show that the compatibility of a filler material with a polymer matrix can be influenced by the particle size of a filling material, where the particle size of a small filler material can increase the degree of polymer strengthening compared to a larger size, the smaller the particle size the higher the bond between the filling material and the polymer matrix (7).

In this research, LDPE nanocomposite-based packaging materials have been made with CB reinforcement material with various compositions using conventional hot and cold press. This is done to determine the proper process parameters or conditions in the manufacture of LDPE nanocomposites and CB which are used as packaging materials that have high thermal resistance and good mechanical properties. To find out the success of LDPE and CB preparation results, physical characterization includes XRD, SEM, and FTIR, mechanical properties including elongation at break, tensile strength, and modulus of elasticity as well as thermal properties including TGA test.

II. Method
The equipment used in this study consisted of a set of glass tools, analytical balance, 200 mesh sieve, ball mill, hot plate & magnetic stirrer, oven, internal mixer, sample mold, cutting machine, Hot and Cold Press Hydraulic, SEM-EDS, FTIR, XRD, UTM, and TGA. The material used is thermoplastic Low-Density Polyethylene (LDPE) produced by PT. Nusantara Petrochemical Interindo, Carbon Black (Carbomax), Aquadest, HCL 37%, and NH₄OH.

LDPE-CB nanocomposite in this study was made by mixing thermoplastic LDPE materials and CB with internal mixers for 10 minutes at 150°C until evenly (homogeneously) with variations in LDPE composition: CB, namely (95: 5, 90:10, 85: 15 and 80:20) %wt. Then put into a square mold with a plate thickness of 0.1 cm, 11 cm long, and 11 cm wide. After that, printing is done by printing hot press for 15 minutes at 150°C printing temperature, followed by cold pressure for 5 minutes at 22°C, then the sample in sheet form is removed from the mold. The printout is then cut to the test sample using a JIS K678 cutting machine. To see the effect of CB addition on LDPE thermoplastic, physical tests (XRD and SEM) were carried out, mechanical tests (elongation at break, tensile strength and modulus young) and thermal test (TGA) on each fabricated nanocomposite.

III. Result and Discussions
The results of x-ray diffraction analysis of CB in Figure 1 shows that the maximum peak diffraction pattern at the inter-lattice angle 2θ = 264.3375° with spacing between fields is d = 1.44680 Å, FWHM = 0.22980 and the peak intensity is 459 a.u.
The results of the x-ray diffraction pattern analysis using Match! Version3 software (Figure 1) obtained the highest peak is the compound that dominates as CB constituent material, C (carbon) which has a rhombohedral crystal structure and orientation of the crystal plane [0 0 21] with lattice parameters \( a = 2.5222 \, \text{Å} \) and \( c = 43.3450 \, \text{Å} \). Whereas small peaks with weak diffraction intensity are CaO (Calcium Oxide) compounds with the orientation value of the crystal field, namely [2 0 0].

From the results of x-ray diffraction peaks, we can obtain the diameter of the CB diameter by using the Scherrer equation:

\[
D = \frac{k \lambda}{\beta \cos \theta}
\]

with \( \beta \), \( r \), \( k \), and \( \lambda \) are the half-peak width (FWHM) in radians, Scherrer constants (0.9), x-ray (1.5406 Å) wavelengths obtained by CB particle size around 36.74 nm. To do the morphological analysis of CB, Scanning Electron Microscope-Energy Dispersion Spectroscopy (SEM-EDS) is used.

Based on the observations in Figure 2 CB shows that the surface morphology is shaped like irregular glass flakes and is classified as amorphous. While the dominating elements are carbon (C) 86.07 %wt, oxygen (O) 13.37 %wt and calcium (Ca) 0.56 %wt (figure 3).
Intermolecular interactions in LDPE-CB nanocomposite formation can be obtained from FTIR testing. The FT-IR test results are obtained in the form of a spectrum that illustrates the magnitude of% Transmittance and wave number for LDPE-CB nanocomposites (Figure 4). In the IR spectra, the absorption peak at wave number 2915.11 - 2847.41 shows stretching vibration absorption for -CH of the alkane. Whereas for absorption peaks at numbers 1462.35-1377.87 shows that there has been an extension of the polymer chain for the asymmetry -CH functional group with CH3. The widening of absorption peaks at numbers 729.25, 719.03, 573.04, 531.14 and 513.46 which shows changes in the shape of -CH groups in mixed solutions. The result of IR spectra analysis shows the appearance of LDPE-CB nanocomposite absorption characteristics and the absence of new groups from the mixture indicates that the results of LDPE-CB nanocomposite interactions in solution are limited to physical bonds.

From the analysis of the combined diffraction pattern data (Figure 5) CB nanocomposite XRD, the diffraction pattern generally follows the pure LDPE diffraction pattern. The addition of CB with the composition (5, 10, 15, 20) % wt only experienced a shift in the intensity of the diffraction peak not too significant. This indicates that the thermoplastic LDPE nanocomposite with CB filler does not have a perfect bond, causing a decrease in tensile strength and elongation at break of nanocomposites due to the CB crystal structure with LDPE.
Figure 5. Comparison of X-ray Diffraction Patterns for LDPE-Carbon Black Nanocomposites of Various Compositions

The morphology of the samples was analyzed using SEM to obtain further information about the effect of adding CB weight composition on the mechanical properties of the resulting LDPE nanocomposites and to see the distribution of fillers in the matrix. LDPE-CB nanocomposite surface morphology results for optimum mix ratio with less optimum.

Figure 6 (a) 95% LDPE morphology and 5% carbon black; (b) 80% LDPE morphology and 20% carbon black

In Figure 6 (a) and Figure 6 (b) there is a thermoplastic LDPE nanocomposite morphology with CB fillers with a composition of 5% and 20%. From the figure, it appears that nanocomposites tend to agglomerate with the addition of composition because CB is hydrophobic which tends to separate with LDPE so the surface energy gets higher.
In Figure 7, it can be seen that with the increase in CB, the elongation of the drop decreases. The tensile strength test results on the CB composition are shown in Figure 8.

In Figure 8 above, it can be seen that increasing CB causes decreased tensile strength. The test results of a modulus of elasticity on CB composition are shown in Figure 9.
Figure 9 shows that increasing CB content causes the modulus of elasticity to increase. The results of mechanical tests on thermoplastic LDPE nanocomposite samples with CB fillers showed tensile strength, elongation and the modulus of elasticity occurred in successive quality decreases ranging from the composition of 5-20%. This is due to the high carbon (C) content and the existence of a lump which is believed to be a place of stress concentration and the initial occurrence of a decrease in strength which results in the nanocomposite being brittle or easily cracked. This is the same as research (8).

The decrease in the tensile strength and elongation of the break is also caused by an increase in the interaction process of poor adhesion force, can be seen from the increase in the pivot that arises and results in poor interaction between materials. The particle size has a relationship to the surface area per gram of filling material. The small particle size produces a large surface area on matrix polymers and fillers so as to enhance the strengthening of polymeric materials. This is consistent with the results of the study (7) which states that the surface area can be increased by the presence of a surface that is a pivot or cavity on the filling surface. Polymers can enter the surface that is shafted during the mixing process. Homogeneous scattered particles increase interaction through absorption of the polymer over the surface of the filler material. Conversely, particles that are not scattered homogeneously may produce lumps in the polymer matrix. The presence of lumps in the polymer matrix reduces the surface area then causes interaction between fillers, the matrix weakens and results in a decrease in the physical properties of polymeric materials (8).

![Figure 10a](image)
![Figure 10b](image)

**Figure 10.** (a) Thermogram 95% LDPE and 5% carbon black; (b) Thermogram 80% LDPE and 20% carbon black

Figure 10a and 10b on LDPE nanocomposite with CB filler material, it can be seen that the composition of 5% with decomposition temperature 488.70 °C changes in mass 96.01% weight, while at composition 20% with a temperature of 490.11 °C changes in mass 91.82% weight. The more temperature changes, the greater the mass change, this is because when the nanocomposite receives heat the matrix undergoes an exothermic reaction, where the nanocomposites emit heat which causes physical changes (deformation) and chemistry which eventually form carbon residues. The inter-particle bonds that occur in nanocomposite materials play an important role in increasing and limiting material properties. Nano-sized particles have a high interaction surface area. The more particles interact, the higher the thermal conductivity. However, the addition of nanoparticles will not always improve thermal properties, there are certain limits which, when added, thermal properties actually decrease. But in general, nanocomposite materials show differences in thermal, mechanical, electrical, optical, electrochemical, catalyst, and structure properties compared to their constituent materials (9).
IV. Conclusions
The effect of adding CB on LDPE thermoplastic was carried out in this study. From the results of the study, it was found that the CB particle size was about 36.74 nm and the dominating compound as a constituent material was C (carbon) which had a rhombohedral crystal structure. The addition of CB to thermoplastic LDPE as a nanocomposite fill material for packaging material applications is able to withstand heat well and is not easily degraded by temperature. This result is supported by the decreasing particle size distribution with increasing thermal properties added and IR spectra which give results that only physical bonds occur. The optimum composition results in CB 5% weight filler where the surface morphology analysis tends to agglomerate, the results of the mechanical analysis showed a tensile strength of 21.45 MPa, elongation at break 436.21 mm, and modulus young 246.22 MPa, and thermal analysis with the decomposition temperature of 488.70 °C changes in mass.

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