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Effect of crude palm oil, *Aloe vera*, glycerol, and starch on characteristics and mechanical properties of polyethylene film

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Abstract

The long-chain structure of polyethylene (PE) have caused low degradation ability of PE, and its hydrophobic nature have reduced its compatibility with other hydrophilic polymer material. This paper focuses on the effect of blending crude palm oil (CPO), *Aloe vera* (AV), glycerol (G), and starch (S) with PE on the characteristics of PE-based film. The films were prepared using melt blending and hot press techniques to obtain a flat sheet film. The FTIR results have shown that the addition of CPO and glycerol reveal the presence of C=O stretching of the ester group in the PE-based film. The addition of CPO, AV, S-G, and S-CPO reduced the degree of crystallinity of PE-based film by disappearance of 2θ = 21.4774 and 23.7334. Furthermore, the addition of CPO, AV, glycerol, and starch increased the thermal degradation rate of PE-based film, where AV shows the best percentage of weight reduced. The crystallinity of the PE-based film has been disrupted, as shown in the XRD analysis. PE-AV has the highest tensile strength, elongation at break, and lowest Young modulus. In conclusion, AV has better potential then CPO in improving the mechanical properties of PE-based films. Therefore, AV has the ability to become a filler in producing packaging film.

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1.0 Introduction

Polyethylene (PE) is the most common petroleum-based synthetic polymer that is being used in the packaging industry (Makhtar et al., 2013). Polyethylene is exceptionally stable and very suitable for packaging applications (Datta & Halder, 2018). The beneficial properties of PE that are cheap, light, excellent processability, less energy consumed, and high resistance to the chemical and harsh environment makes PE as the first choice for the manufacturer (Ratnam et al., 2006; Sabetzadeh et al., 2018). However, PE has low degradation because of its chemical structure which is unable for microorganisms to attack for degradation (Al-Salem et al, 2019; Datta & Halder, 2019). Hence, the development of degradable PE should be explored.

Plastic based film packaging covers one-third of sales in the world (Datta & Halder, 2018). It is primarily used in the food packaging industry (Jaafar et al., 2018; Majid et al., 2016). The bio-based polymer has biodegradable properties. Polyhydroalkanoates, polylactide (PLA), PLA blends, and starch blends are among the most common bio-based polymer materials used to produce food film packaging. However, their applications are limited due to their poor strength and processability (Hejna et al., 2019; Nguyen et al., 2016). Consequently, more research should be done to enhance their properties. One of the studies which can be explored is by combining PE and bio-based polymer.

A combination of PE and starch is a combination of synthetic and bio-based polymer, which is also known as a partially degradable polymer. Starch is cheap, biodegradable, compostable, high availability, and has excellent film-forming ability (Gutiérrez & González, 2017; Linares et al., 2019; Mazerolles et al., 2019; Pinzon et al., 2018). Nonetheless, starch films have limited applications as a component in polymer blends (Hejna et al., 2019). Starch is highly sensitive to moisture due to hydrophilic nature (water sensitivity), which weaken the mechanical properties (Ahmadi et al., 2018; Martins et al., 2018) and reduce the barrier properties of the film (Gutiérrez & González, 2017). Therefore, an addition of plasticizer will disrupt the semi-crystalline of starch to improve starch properties (Martins et al., 2018).

Plasticizers were used to enhance the flexibility and processability of polymer, either synthetic or bio-based. Common bio-based plasticizers are epoxidized soybean oil, epoxidized linseed oil, castor oil, palm oil, glycerol, vegetable oils, sugars, and starches. The bio-based plasticizer was introduced to replace synthetic plasticizer because it has lower toxicity (Kamarudin et al., 2019). Kamarudin et al., (2019) modified polypropylene (PP) with crude palm oil (CPO), which leads to more free volume and enable the network and proved the occurrence of plasticization. Epoxidized palm oil and epoxidized soybean oil show molecular interaction with PLA and improve the plasticization effect in PLA (Tee et al., 2014). Glycerol had been used in many starch-based films because its hydrophilic compound that able to enhance mechanical properties of the film (Vieira et al., 2011).

The crosslinker is typically used in polyethylene/starch (PE/S) based film to improve the mechanical properties of the film, which is also known as compatibilizer. Common cross-linker or compatibilizer used in PE/S film is polyethylene maleic anhydride (PE-g-MA) (Ahmadi et al., 2018; Nguyen et al., 2016; Sabetzadeh et al., 2017). PE-g-MA is a synthetic compatibilizer that seems promising, but the mechanical properties remained unsatisfying and inconsistent (Datta & Halder, 2018; Nguyen et al., 2016; Sabetzadeh et al., 2017). Therefore, other researchers add other crosslinkers such as nanoclay (Sabetzadeh et al., 2016), and zeolites (Thipmanee et al., 2015).

This research used *Aloe vera* (AV) gel as the crosslinker. AV gel had been established in the pharmaceutical and cosmetic industry. In the food packaging industry, the presence of organic acid mainly citric acid in AV able to give a cross-linking effect between starches in the film and improve the mechanical properties of the film (Gutiérrez & González, 2017). AV also had been incorporated in carboxymethyl cellulose-polyvinyl alcohol film and become an active component in preventing photo-oxidation (Kanatt & Makwana, 2020). Another study found the solubility, antioxidant and tensile strength of the combination of extra virgin olive oil with chitosan improved when AV was added into the formula (Pratama et al., 2019).

This current research investigated the impact of AV, CPO, glycerol (G) and starch (S) towards PE-based film. The films were prepared using melt blending and hot press technique. Characterisation of the film was conducted as to determine the presence of the functional group, the changes in the glass transition temperature, the degradation temperature profile, the appearance of the crystalline structure, and the performance of mechanical properties.

2.0 Methodology

2.1 Materials

The materials used in this study were low-density polyethylene (MW:0.915 g/cm^3, Lotte Chemical, Titans, Malaysia), potato soluble starch (Bendosen, Malaysia), glycerol (MW:92.09, Chemiz, Malaysia), crude palm oil (4.00 free fatty acid, Malaysia Palm Oil Berhad, Malaysia) and, extracted AV gel (Aloe Barbadensis, Malaysia).

2.2 Methods extraction of Aloe vera gel

The modification method from (Gutiérrez & Álvarez, 2016; Pinzon et al., 2018) was applied. AV leaves were kept for 15 minutes as to allow the latex to drain off. The leaves were then washed with distilled water to remove any impurities before paper towel dried. The outer green skin of the leaves was then removed, and the AV parenchyma (AV gel) was collected. The gel was mashed using a food blender for 3 minutes before filtered using cheesecloth to discard the impurities (residue). The filtrate (AV gel) was used as a cross-linking agent in this study.

2.3 Preparation of the film

Melt blending and hot press technique were applied in the development of the film (Gutiérrez & Álvarez, 2016; Kamarudin et al., 2019; Ratnam et al., 2006). Firstly, S and PE were dried in an oven for 24 hours. Next, PE, S, CPO, and AV gel were pre-mixed for 30 minutes and left for 24 hours (in a desiccator). The formulation used was as shown in Table 1. Secondly, the melt blending process was conducted using Banbury mixer brand Thermo Haake Polylab Internal Mixer. The operating condition was 170 °C, 60 rpm for 5–15 minutes. The mixture was then crushed in Rexmac Compact Crusher to a bead size of less than 2 mm. The beads were then hot-pressed using Cometech hot press machine. The beads were placed on a steel plate and pre-heated for 10 minutes before hot-pressed at 130 °C, 700 psi for 10 minutes. The hot film was cooled down by cooling water until it reaches 35 °C. Finally, the film was peeled off from the steel plate. The hot press method was adapted from (Datta et al., 2019).

Table 1: Samples formulation

Sample Name	The percentage ratio of the material
PE	100
PE-S-G	70:20:10
PE-S-CPO	70:20:10
PE-AV	95:5
PE-CPO	95:5

2.4 Characterisation of the film

2.4.1 Spectroscopy

Fourier transform infrared spectroscopy (FTIR) was used for component identification and identification of interfacial interaction of hydrogen bonding between the materials. The measurements were carried out using a Mettler Toledo FTIR spectrophotometer. The range of spectrum was between 400–4000 cm^{-1}, 4 cm^{-1} resolution, and 64 scanning times.

2.4.2 Melting and Crystallisation Behaviour

A Mettler Toledo differential scanning calorimetry technique (DSC) measures heat flow as a function of temperature and time within a controlled atmosphere to study the thermal behaviour. Samples were cut and prepared in encapsulated aluminium pans at a weight of 5.2–5.5 mg. The scans were performed by cycle process; 1) heating samples (25 to 190 °C), 2) cooling samples (190 to −10 °C), and 3) heating samples (−10 to 190 °C). The heating rate of the samples is 20 °C/min under flowrate of nitrogen atmosphere 50 ml/min. The formula used to obtain X_c as per Eq.(1).

$$X_C\ (\%) = \frac{\Delta H_m}{\Delta H_m^o} \times \frac{100}{x_{PE}} \qquad (1)$$

where: X_c is crystallinity percentage, ΔH_m is melting enthalpy or enthalpy of fusion, ΔH_m^o is formation enthalpy or enthalpy of crystallisation, and X_{PE} is PE weight of the fraction.

2.4.3 Thermal Stability

Thermogravimetric analysis was performed using TGA Instrument (TGA/SDTA581, Mettler Toledo) by determining the weight changes as a function of temperature and time. Samples approximately 20 mg were subjected to TGA with operating condition setup for TGA instrument were heating rate at 10 °C/min, the flow rate of nitrogen atmosphere was 50 mL/min, and the temperature range between 25 to 600 °C. Eq. (2) represented the formula to get the weight loss.

$$Weight\ loss\ (\%) = \frac{W_i - W_f}{W_i} \times 100 \qquad (2)$$

where: W_i is initial weight, and W_f is the final weight

2.4.4 Crystallinity

The X-ray diffraction (XRD) brand PANalytical Aeris were used to determine the crystallinity of each sample. The range of diffraction angles (2θ) was 5–70, and the scanning rate was 2.1786×10^{-4} 2θ/s for 25 minutes.

2.4.5 Mechanical Properties

Instron 3382, Universal Testing Machine was used to find the mechanical properties of the samples. The specimens were cut based on ASTM D882 with a length of 100 mm and width 25 mm. The films will be elongated at a rate of 10 mm/min at room temperature.

3.0 Results and discussion

3.1 Spectroscopy

Fig. 1 shows the FTIR spectra for the wavelength between 500 to 4000 cm^{-1}. The peaks obtained were the same for all films at the peak of 719 cm^{-1} and 730 cm^{-1}. The peak at 719 cm^{-1} represents the strong appearance of 1,3 di-substituted C–H bending while 730 cm^{-1} represents the strong appearance of 1,2 di-substituted C–H bending.

Peak 1376 cm^{-1} was demonstrated by each film that represents weak O-H bending. As can be seen, the addition of AV does not have a significant impact on the functional group of the PE film. The peak of 1465 cm^{-1} and 1472 cm^{-1} can be seen for all films representing a medium C–H bending for alkane functional groups. A new peak was observed (1743 cm^{-1}) when CPO and glycerol were added into the film. The peak between 1739–1743 cm^{-1} representing the existence of C=O stretching for the ester group.

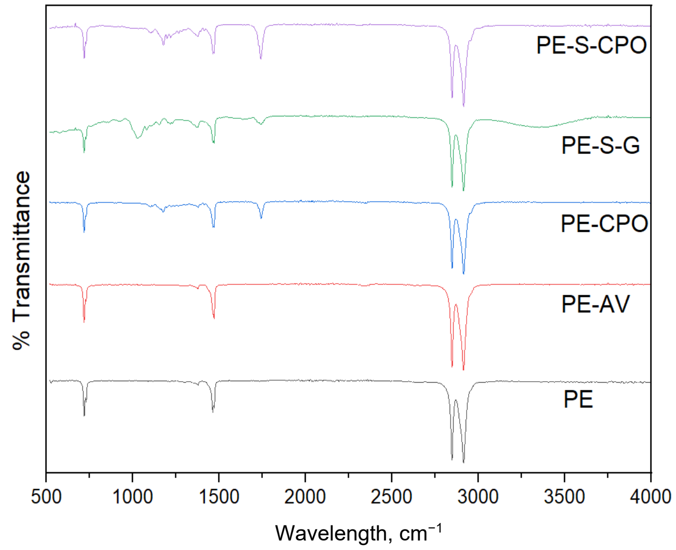

Fig. 1: FTIR result in range of 500–4000 cm^{-1}

This peak indicates the strong C=O bond contributed by triglyceride ester in the crude palm oil (Rahman, 2013). A similar finding was obtained when the PLA mix with epoxidized jatropha oil reveal the presence of C=O stretching of ester at a wavelength of 1741 cm^{-1} (Chieng et al., 2014). This peak had also occurred when glycerol was added into PE-S film; a weak and broad stretching might occur due to the plasticization effect. The presence of new peaks indicates the possibility of interaction between PE-CPO and PE-S with glycerol or CPO. Plasticization effect on PE-S-CPO was higher than PE-CPO. On the other hand, the plasticization effect on PE-S-CPO was higher than PE-S-G because CPO not only acts as a plasticizer for starch but also for PE. Hence, CPO is better plasticizer then glycerol for PE-S film. CPO was able to act as a plasticizer for PE and starch (Emiliana et al., 2012a; Makhtar et al., 2013) while glycerol works as a plasticizer for starch only. Peak 2850 cm^{-1} and 2916 cm^{-1} represent strong O–H stretching of the intermolecular bond of alcohol and medium C–H stretching of the alkane functional group.

3.2 Melting and Crystallisation Behaviour

Fig. 2a and 2b display the melting behaviour (heating curve obtained from the second heating scan) and crystallisation behaviour (cooling scan) from DSC. The glass transition temperature (T_g) was reduced with the addition of CPO, AV, and glycerol. Based on Fig. 2a, only one endothermic peak appeared in all films with a slightly different melting peak temperature. The melting temperature (T_m) detected were very close to each other, as tabulated in Table 2.

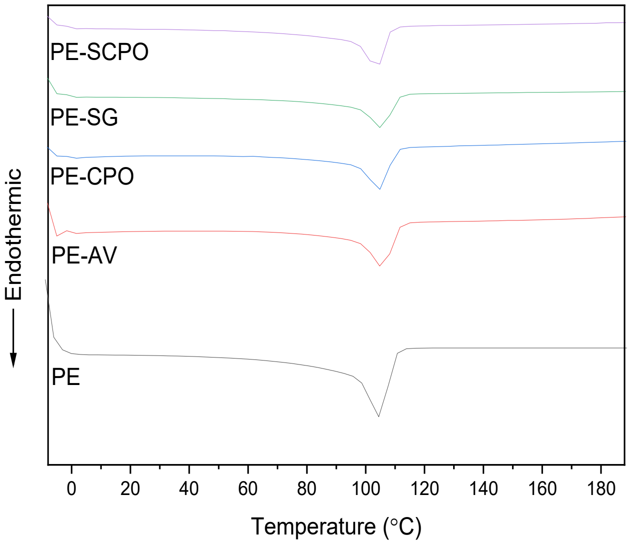

Fig 2a: DSC curve of heating

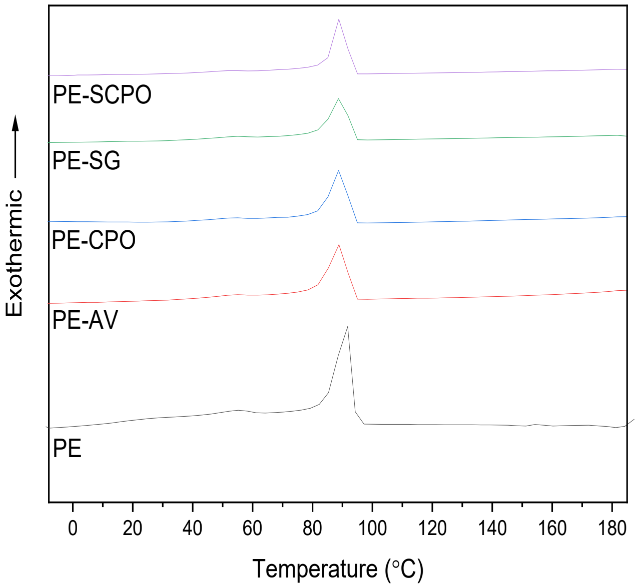

Fig 2b: DSC curve of cooling

Table 2: DSC analysis data

Sample Film	T_g (°C)	T_c (°C)	T_m (°C)	ΔH_m (J/g)	X_c (%)
PE	49.18	91.76	104.44	96.53	32.95
PE-AV	48.39	88.81	104.74	64.25	23.08
PE-CPO	48.34	88.70	104.81	56.48	20.29
PE-S-G	48.37	88.67	104.79	55.79	27.20
PE-S-CPO	48.35	88.76	104.77	48.56	23.68

T_g (glass transition temperature), T_c (crystallisation temperature), T_m (melting temperature), H_m (enthalpy of fusion), X_c (degree of crystallinity)

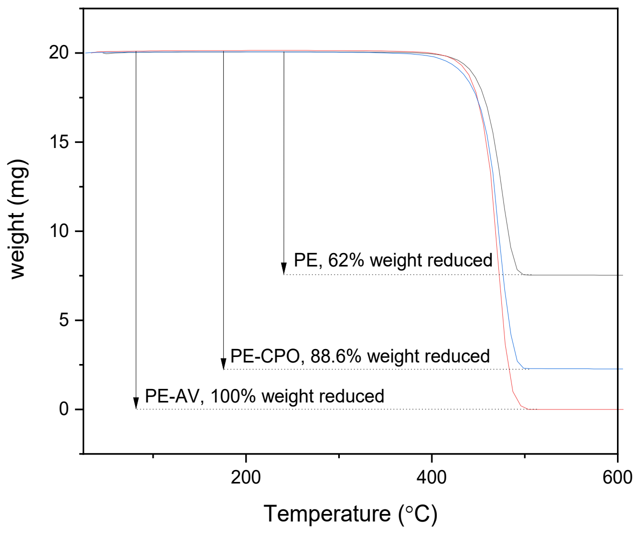

Fig. 3a: Thermogravimetric curve of PE, PE-AV, and PE-CPO

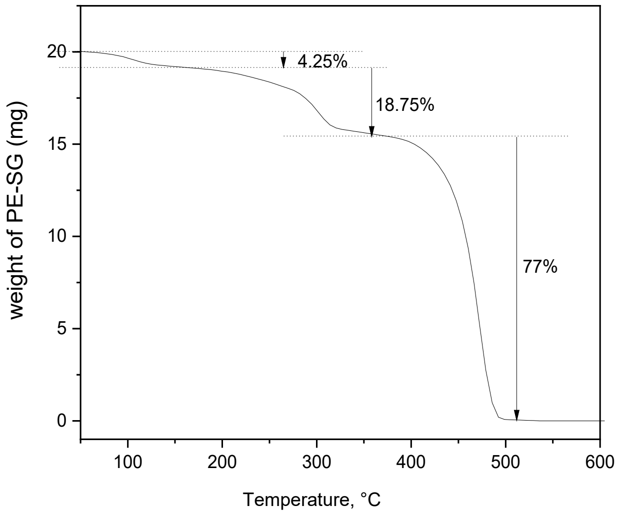

Fig. 3b: Thermogravimetric curve of PE-SG

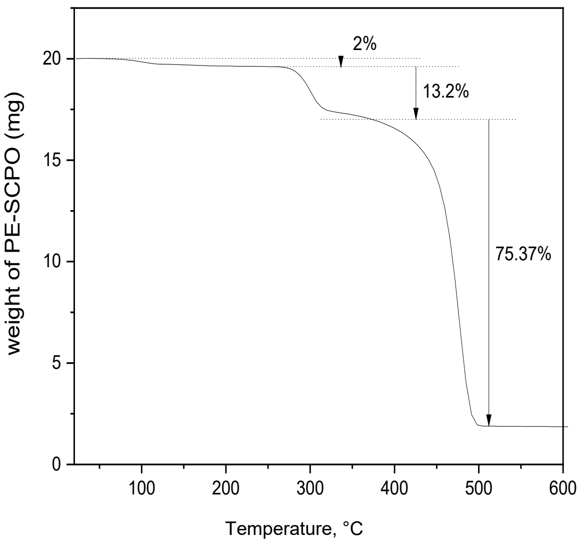

Fig. 3c: Thermogravimetric curve of PE-SCPO

The crystallisation temperature (T_c) of the films had been revealed in the DSC cooling curve, as in Fig. 2b, represented by only one exothermic peak. The peak intensity represents the crystallisation behaviour of the film. It shows that CPO and AV had modified the crystallisation structure of PE film, while glycerol and CPO on the PE-S film. From Table 2, the T_c decreases with the presence of plasticizer in PE and PE-S film. The degree of crystallinity (X_c) for PE film decreased with the addition of CPO, AV, S-G, and S-CPO. CPO and AV are believed to act as nucleation sites for the crystallisation and demolished the crystalline structure of PE. Kamarudin et al. (2019) suggested that CPO and gives a lubricating effect on PE and reduces the X_c. The crystallinity of PE-AV reduced because the amount of PE was lower compared to PE film. Similar result was obtained by other researcher too (Korol et al., 2015). However, based on the results, the X_c value of PE-S-G is higher compared to PE-S-CPO, where the existence of glycerol gives more intermolecular interaction with starch if compared to CPO to starch. The presence glycerol, and CPO in PE-S film respectively had reduced the molecular weight of the starch chain and improve the free volume of the film (Martins et al., 2018).

3.3 Thermal stability

The thermogravimetric analysis (TG) of the films were shown in Figure 3a-3c. Figure 3a shows the TG curve for PE, PE-AV and PE-CPO. It shows that one stage of degradation occurred during thermal degradation analysis. The total percentage of degradation for each film was 62.3% for PE, 88.6% for PE-CPO and 100% for PE-AV. The weight loss percentage of PE increases with the addition of CPO and AV. CPO and AV have disrupted the structure of original PE by reducing the interfacial adhesion, which leads to lowering the interfacial tension and increasing the thermal degradation. The one-step degradation process also occurred due to the addition of CPO in PP (Kamarudin et al., 2019).

Figure 3b and 3c represent the TG curve for PE-S-G and PE-S-CPO. With the presence of starch in PE, three stages of degradation obtained. The presence of starch in both films shows evaporation of moisture content for temperature less than 200 °C (Makhtar et al., 2013). The percentage of weight loss for the first stage is 4.25% for PE-S-G and 2% for PE-S-CPO. Degradation of the second stage shown in PE with the presence of starch because starch degrades between 200 °C to 350 °C. For PE-S-G, it shows the evaporation of G-S content and similar result found by (Gutiérrez & Álvarez, 2016) and evaporation of CPO-S content for PE-S-CPO. The last shift, at around 466.58 °C, is caused by the thermal decomposition of LDPE.

Fig. 3d shows the maximum decomposition rates of the films that were analysed using DTG thermograms. PE, PE-AV, and PE-CPO revealed only 1 degradation step involved in the thermal degradation rate. PE-S-G and PE-S-CPO exposed three levels of degradation rate, which at 60 – 160, 200 – 330, and above 390 °C. The maximum temperature, Tmax for the decomposition of all films were; PE, 478.32 °C, PE-AV, 470.93 °C, PE-CPO 471.91 °C, PE-S-G, 472.62 °C, and PE-S-CPO, 478.38 °C.

The first peak between 60 – 140 °C represents the moisture content evaporated from the presence of S in PE-S-G and PE-S-CPO. Usually, between 0 – 160 °C correspond to water evaporation (Datta et al., 2019). Started from 200 to 330 °C, the DTG thermograms showed degradation of G and S. The presence of S in PE had reduced the thermal stability of the films (Mao et al., 2016). The evaporation of G was expressed by the loss of the volatile components at this temperature.

The decomposition temperature range of S in PE-S-G and PE-S-CPO was 280 °C to 330 °C, depending on the amylose and amylopectin presence in selected starch. The amylose and amylopectin chain and cellulose network structure affecting the decomposition temperature (Amin et al., 2014). The third stage demonstrated the decomposition of PE started at 390 °C.

3.4 *Crystallinity*

Fig. 4 represents the XRD patterns of the films. The neat PE shows two main peaks at around 2θ = 21.4774 and 2θ = 23.7334, attributed to the partially crystalline formation of PE. Similar findings obtained by (Alnaimi et al., 2015). The intensity of pure PE peaks decreases on the addition of other materials, and new peak observed at around 2θ = 44.5, 2θ = 44.0, 2θ = 51.5 and 2θ = 61 – 62. According to (Domene-López et al., 2019), the original peak of S-G was at 2θ = 17.1, 19.7, and 21.5. For this reason, S-CPO was expected having the same characteristic as S-G mainly due to CPO act as plasticizer same as glycerol. The characteristics of AV show a completely amorphous pattern (Ortega-Toro et al., 2017). It may be inferred that S-G, S-CPO, and AV have successfully immobilized in PE and showed an almost same new peak of crystallinity. For PE-CPO, the CPO reacts as a plasticizer and turns the crystalline structure of PE into the amorphous phase. However, the results contradicted with the degree of crystallinity obtained from the DSC result. The intermolecular hydrogen bonding being disturbed by other materials (AV, CPO, S-G, and S-CPO), leading to loss of crystallinity structure PE.

3.5 *Mechanical Properties*

The stress-strain curve of PE film with CPO and AV and PE-S based film with CPO and G as the plasticizer are presented as in Fig. 5. As seen from Fig. 5, the rising of the stress is linear, with the strain at low strain for all materials. Then, the stress exhibits constant stress until the film break at higher tension.

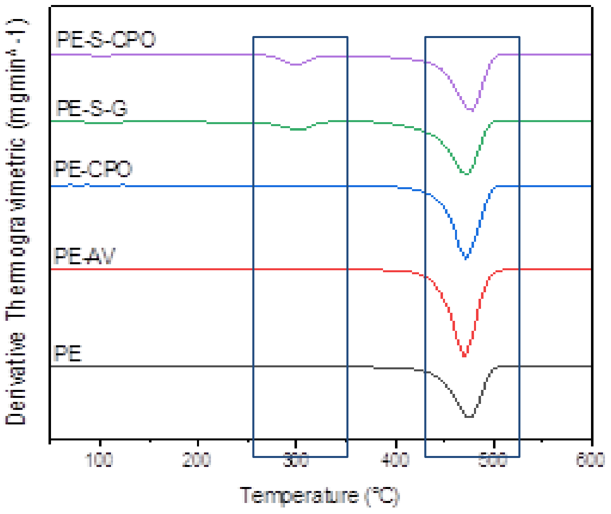

Fig. 3d: Derivative thermogravimetric curves of PE, PE-S-G, PE-S-CPO, PE-AV, and PE-CPO

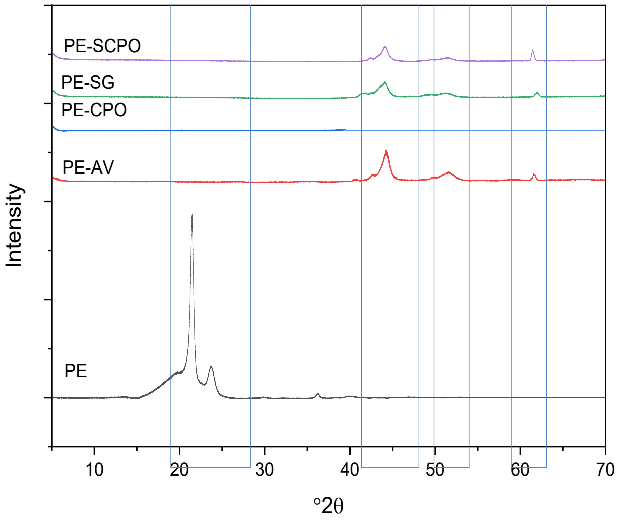

Fig. 4: XRD pattern of PE, PE-S-G, PE-S-CPO, PE-AV, and PE-CPO

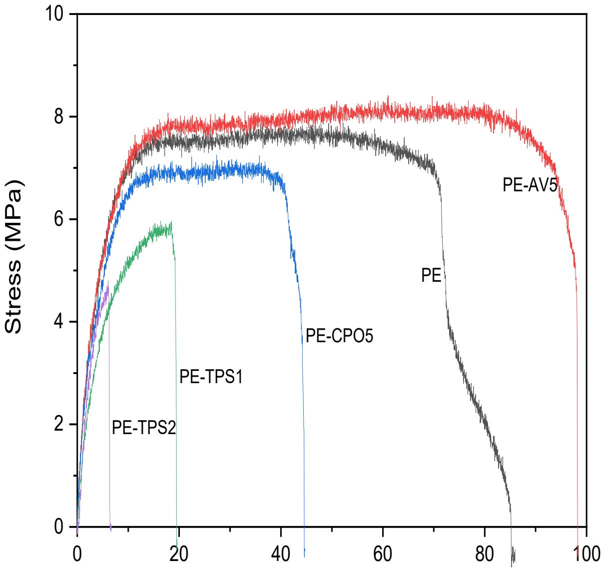

Fig 5: Stress-strain curve

Table 3: Mechanical Properties of the films

Sample film	Tensile Strength (MPa)	Elongation at Break (%)	Young Modulus (MPa)
PE	7.79	35.40	129.93
PE-AV	8.31	57.87	84.75
PE-CPO	7.06	21.60	120.42
PE-S-G	5.84	16.79	89.20
PE-S-CPO	4.85	6.15	156.67

The presence of starch in PE-S-G and PE-S-CPO had caused a brittle fracture that occurred at low extension.

The function incorporating plasticizer into PE and PE-S based film was to improve flexibility and prevent the product from breaking easily. Table 3 presents the value of tensile strength, elongation at break, and young modulus of the film. The tensile strength and elongation of PE improved with the addition of AV into PE film while the addition of CPO reduced the tensile strength and EAB of PE. The presence of AV in the film had cause plasticization effect occurred and increase the elongation at break of the film. AV has the possibility to act as a cross-linker that improves the cross-linking between PE molecules. The tensile strength (TS) result is supported by the X_c. Higher X_c value gave better TS value because of greater packing chain and chain structure (Ratnam et al., 2006). Young's modulus represents the stiffness of the material that shows the relationship between stress and strain. PE-AV5 has the lowest stiffness. For PE-starch film, glycerol gives better TS, EAB, and YM to the film instead of CPO. Glycerol was giving a positive stretching effect to the PE-starch film and similar result obtained by (Sabetzadeh et al., 2015). CPO gives a lubricating effect and reduces the cohesion forces that lead to slip and slide off the molecules easily. This situation increases the free volume in PE and PE-starch network structure and finally weakens the interaction between the polymer chain. Similar findings on the lubricating effects found on the combination of CPO and polypropylene by the previous researcher (Emiliana et al., 2012b).

4.0 Conclusions

PE and PE-starch based films with a different type of plasticizer have been extensively synthesized using melt blending and hot press technique. PE-AV film was found to produce better tensile strength, elongation at break and young modulus compared to PE and PE-CPO. For PE-S film, the presence of glycerol improved the compatibility between PE and starch by providing better tensile strength, elongation at break and young modulus compared to the addition of CPO. AV gel can be proposed as potential additives to improve PE film mechanical properties.

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