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The Plasticization Effect of Hydroxylated Palm Stearin Plasticizer (HPS) Towards High-Density Polyethylene (HDPE) Via Compression Moulding

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Abstract

Hydroxylated palm stearin (HPS), a chemically modified palm oil derivative, was investigated as a bio-based plasticizer for high-density polyethylene (HDPE). HDPE was compounded with 10, 20, and 30 wt.% HPS, and the plasticization behaviour was evaluated using mixing torque, tensile testing, morphology, thermal properties and chemical interaction. This finding revealed that HPS significantly enhances the melt processability of the resin by reducing the mixing torque by 79.2%. The increment of the HPS content tends to reduce the tensile strength of the blends up to 46.5% due to enhance HDPE chain mobility. The morphology observations revealed the presence of dark voids, confirming the plasticization effect happened towards HDPE. DSC analysis showed a slight reduction in melting temperature and an increase in crystallization temperature, while the degree of crystallinity remained largely unchanged. HDPE/HPS also demonstrated a gradual reduction in thermal stability, with the degradation temperature ($T_{d5\%}$) decreasing from 413.0 °C (HDPE) to 280.6 °C (70/30). FTIR analysis confirmed that no new chemical bonds were formed during blending, indicating that HPS acted as a non-reactive plasticizer through physical interactions. These findings showed HPS effectively improves the processability of HDPE while maintaining its crystalline structure, although excessive HPS loading compromises the mechanical properties.

Keywords

Polymer Blend; Compression Moulding; Hydroxylated Palm Stearin (HPS); Plasticizer; High-Density Polyethylene (HDPE);

1. Introduction

The global plastic waste crisis is a multifaceted issue driven by various socio-economic factors. Mainly, the plastic waste accumulation is driven by the continuous plastic product demands and consumption of plastics which particularly depends on the plastic application which is single-use packaging and inefficient solid waste management. Global plastic production was projected to reach 500 million metric tons in 2025, with expected to surpass 12 billion metric tons by 2050 [1]. The reliance on single-use plastics is heavily influenced by the consumer and industrial habits. Plus, the inefficiency of waste management systems such as open dumping and landfilling had caused global landfill demand for the waste disposal [2, 3].

Considering that, a lot of study was done to replace the synthetic-based plastic with biodegradable materials, to produce biodegradable or biohybrid blends polymer. Somehow, biodegradable materials also have some minor issues such as strength limitations, microplastic issues and sensitive storage conditions due to moisture and heat sensitive materials [4, 5]. So, this study was to show a new biohybrid blend made from high-density polyethylene (HDPE) with the presence of newly synthesized hydroxylated palm stearin (HPS) as a novel plasticizer.

In previous few years, palm oil-based plasticizer was utilized in HDPE blend. Crude palm oil (CPO) showed the possibility to be used as biodegradable plasticizer in the HDPE blend [6, 7]. It was shown that a small amount of CPO (2-5 wt%) improved the tensile strength of the HDPE from 25 MPa up to 32 MPa [6]. Plus, the surface morphology showed a high extent of plastic deformation at 500% elongation [6]. However, previous study only focused on the small amount of palm oil-based plasticizers which is up to 5wt%. On the other hand, the palm oil-based plasticizer was possible to be blended with other polymer, which is PLA with the content up to 20wt% [8]. As referred to the Wypych et al. [9], the ability of plasticizer applications could utilize up to 50wt% and depending on the plasticizer's characteristics.

This study was expected to show the ability of the HPS to plasticize the HDPE while giving a good characteristic of a biohybrid resin which was maintained the high tensile strength while reflecting a good thermal degradation property towards biodegradability. Palm stearin was suggested since it has higher melting points and more rigid characteristics compared to the crude palm oil [10] which is suitable for HDPE blend as plasticizer component. Apart from that, the use of palm stearin as a plasticizer in the HDPE blend is not fully discovered. Besides HDPE, palm stearin already been used as plasticizer in PLA and PBAT systems [11–13] which here is a possibility where the palm stearin could be used as plasticizer in HDPE blend. By the introduction of hydroxylation process, a new synthesis process was done towards palm stearin [14] to form a new novel plasticizer known as hydroxylated palm stearin (HPS). Plus, with the high tensile strength of the HDPE, it was expected that the application of the HPS could be more than 10wt% in the HDPE blend.

Thus, the current study aimed to bridge the gap between the limitations of palm stearin as the plasticizer in the HDPE blend by observing the plasticization behaviour of the HPS towards HDPE. HDPE was blended with 10, 20 and 30 wt% of the HPS using an internal mixer and the effects discussed were tensile properties, morphological, thermal properties and chemical interactions from FTIR.

2. Methodology

2.1 Materials

High-density Polyethylene (HDPE), (Grade Name: HD5301AA) obtained from Lotte Chemical Titan. The hydroxylated palm stearin (HPS) was synthesized from (refined, bleached and deodorized (RBD) palm stearin supplied by FGV Holding Bhd. and was beaded into small granule-powder form.

2.2 Material Compounding

A set range of formulation was tabulated in Table 1 by varying the HPS and compatibilizer alternately to maximize the observation on blending effects. Initially, the HDPE pellet was manually inserted into the internal mixer (Brand: Haake Rheomix OS Thermo Scientific). The mixing parameter was set at the temperature of 180 °C and screw speed of 5 rpm. The mixing speed gradually increased up to 50 rpm before the HPS was added slowly during mixing process. The mixing time of the blends were measured until the blends achieved stabilized torque value.

Table 1 Formulation of HDPE-HPS Blends

Sample Category	Sample ID	HDPE (%)	HPS (%)
HDPE	100/0	100	-
	90/10	90	10
HDPE-HPS	80/20	80	20
	70/30	70	30



Fig. 1 Sample Image of Mixed Resin from The Internal Mixer

After mixing process, the sample was directly cut into small pieces (Fig. 1) before crushed into a fine coarse or granule by using a compact crusher machine. The crushed sample can be referred in Fig. 2.



Fig. 2 Sample Image of Crushed Resin of HDPE-HPS Blends

2.3 Compression Moulding

The 50-tonne hot press machine (Model: Cometech QC602A) was preheated to 30 °C for 3-5 minutes. The compression moulding process consisted of 7 minutes preheating, 10 minutes of heating at the temperature of 150 °C and followed by water cooling process with setting temperature of 50 °C for 10 minutes. During the process, the pressure applied was 25 kg/cm². The arrangement of the metal plate and mould position as shown in Fig. 3.

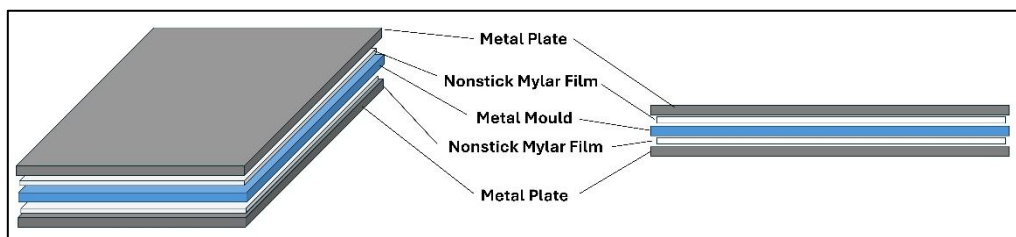


Fig. 3 Arrangement of Plate position (Oblique View and Side View)

2.4 Characterization

2.4.1 Tensile Properties

The sample's shape and size were prepared according to the ASTM D638 Type-I standard. 10 mm /min and 10kN were the crosshead speed and load cell, respectively for the tensile property's parameter setup of the machine, which referred to the standard parameter testing for plain and reinforced polymers [15]. The sample was placed on the gripped area of the universal tensile machine, UTM (Model: Tinius Olsen H50KT) and the sample was pulled while the stress from the sample recorded along with change in strain and automatically plotted in a stress-strain diagram until the sample breakage. The specimen's image can be referred to Fig. 4.



Fig. 4 HDPE-HPS Blends Specimen Following the ASTM D638 Type-I

2.4.2 Scanning Electron Microscopy (SEM)

This analysis was included to obtain the surface structure condition when the HDPE matrix was modified with HPS plasticizer and additives. The fractured samples of HDPE-HPS blends were prepared by cutting the sample according to the size of 4 mm wide and 4 mm long. The sample was turned to the side to observe the cross-section of the resin blend. All samples were coated with platinum using a vacuum coating machine until all surfaces were coated to ensure electron beam can deflect efficiently and giving clear scanning image. Then, the observation was carried out with a parameter voltage of 1.5 kV [16]. The suggested magnification for this analysis was 500x magnification.

2.4.3 Differential Scanning Calorimetry (DSC)

The Differential Scanning Calorimetry (DSC) analysis aimed to ascertain the glass transition temperature (T_g) and melting state and behaviour of the sample blended formulation. Preheating from 30 °C to 200 °C and slow cooling was promptly to eliminate the thermal history. The cooling process was done with cooling rate (10 °C/min) from 200 °C until -160 °C. Then, the samples were slowly heated from -160 °C to 200 °C at a heating rate of 10 °C/min to obtain actual specimens' melting behaviour.

The crystallization characteristics of the HDPE/HPS blend were analyzed by using melting enthalpy of the blends. The degree of crystallinity ($X_c\%$) for each blend was calculated based on the equation below:

$$X_c\% = \frac{\Delta H_m}{\Delta H_m^0 \times w} \times 100$$

Where, ΔH_m is the melting enthalpy of the blend obtained from the DSC analysis, ΔH_m^0 represents the fusion enthalpy of fully crystalline HDPE equivalent to 293 J/g and w represents the weight fraction of the HDPE from overall mass of the blend [17].

2.4.4 Thermogravimetric Analysis (TGA)

Thermogravimetric analysis (TGA) was employed to assess the thermal stability of the samples based on their respective formulations. The sample was weighed in range of 10 to 15 mg and placed in a crucible. The testing temperature was set from 30 °C to 600 °C. A consistent heating rate of 10 °C/min was employed to monitor the sample's mass variation throughout time. The nitrogen purging rate was set at 50 ml/min.

2.4.5 Fourier Transform Infrared Spectroscopy (FTIR)

This characterization analyzed the composition of the samples, including the properties of chemical bonds in the sample blending. The wavelength range should be between 4000 and 400 cm^{-1} with a resolution of 4 cm^{-1} [18]. The scan number of the analysis was 16 scans. With the addition of HPS in the formulation, a few changes on the spectrum peaks were expected compared to the virgin HDPE spectra that represents the chemical bond of the material blending.

3. Results and Discussions

3.1 Mixing Torque Analysis

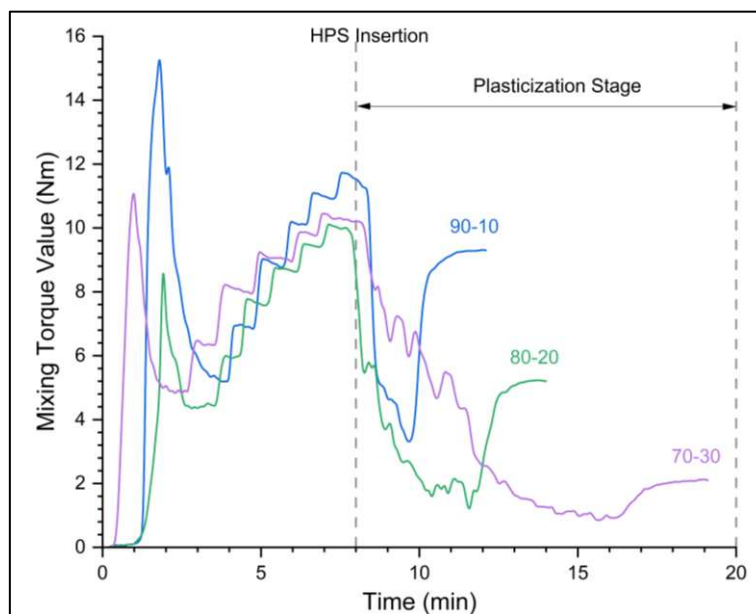


Fig. 5 Mixing Torque Curve of the HDPE-HPS Compounding Process

Fig. 5 showed the mixing torque curve during HDPE/HPS blending occurs across different range of HPS amounts (10-30 wt.%). The mixing process was started by inserting the HDPE into the mixing chamber at 5 rpm. A sharp torque peak is observed within the first 2 minutes, which corresponds to the initial resistance of the solid HDPE pellets as they are loaded into the mixer and begin to melt under the mechanical shear of the mixing screw at 180 °C.

Then, the screw speed was gradually increasing the rpm up to 50 rpm between 3rd and 8th minutes, where the torque had kept stabilized as the HDPE reaches a uniform melt state, reflecting the baseline viscosity of the polymer matrix.

As the mixer achieved 50 rpm at the 8th minutes, the HPS was slowly inserted until the HDPE and HPS blended well. Upon the insertion of HPS plasticizer at the 8-minute mark, there is an immediate and significant drop in torque across all formulations (90/10, 80/20, and 70/30). As shown in Fig. 5, the mixing torque of the blends was reduced as the amount of HPS increased which was from 20.9% for the specimen 90/10 up to 79.2% for the specimen 70/30. This demonstrates that the melt flowability and processability of HDPE increased as the amount of HPS plasticizer increased. In addition, there was no chemical reaction occurred after the homogenous mixing achieved between HDPE and HPS as can be seen in the Fig. 5. This reduction is a clear indicator of the plasticizing effect, where HPS molecules diffuse into the HDPE matrix, reducing polymer-polymer interactions and increasing the free volume. As mentioned by previous author, the impact of plasticizer towards HDPE mixing torque was due to plasticizer act as internal lubricants which resulting to improving molecular mobility and flow of the materials [19, 20]. The occurrence of this behaviour happened by the diffusion of the plasticizer into the polymer matrix [9], where it reduces the polymer-polymer interactions and increasing the free volume [21]. So, the change of these properties lowered the viscosity and mixing torque value [22]. Plus, the plasticization effect shown in the torque analysis could be supported by the result shown in the mechanical properties where the tensile strength (Fig. 6) of the specimen reduced as the amount of the HPS increased while still maintaining elongation at break (Fig. 7) as the HPS applied in specimens 90/10, 80/20 and 70/30.

However, the increment of HPS content to HDPE increased the mixing time during the compounding process. The mixing time for each specimen increased gradually from 12 min, 14 min and 19 min for the sample 90/10, 80/20, and 70/30, respectively. This situation possibly occurred due to the reduction of the torque value as the amount of HPS increased, lowering the torque resistance of the materials towards the mixer's screw. Thus, it requires longer mixing time for the blend 70/30 to achieve the stabilized torque value and homogeneous mixing in comparison to 90/10 blend.

3.2 Tensile Properties

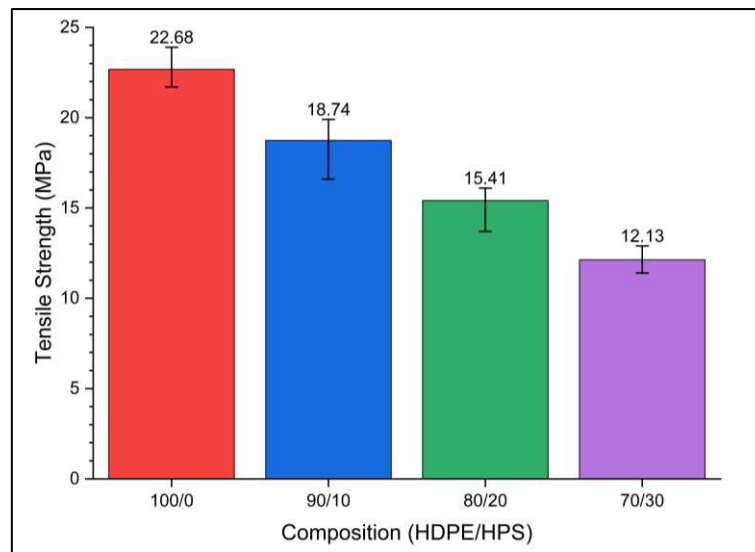


Fig. 6 Tensile Strength of HDPE/HPS Blends

Fig. 6 shows the tensile strength variation based on the composition of HDPE/HPS blends. It showed a reduction of tensile strength value of the HDPE from 22.68 MPa to 12.13 MPa after 30 wt.% of HPS was added. The percentage reduction of the tensile strength of HDPE/HPS blends by comparing to specimen 100/0 was 17.4%, 32.1% and 46.5% for the specimen 90/10, 80/20 and 70/30, respectively. It is expected that the tensile strength shall be reduced due to the interruption of HPS to intermolecular bond HDPE. As the amount of HPS increased, the tensile strength reduced as the intermolecular forces weakened by the increment of plasticizer content. In addition, the reduction possibly occurred due to the increment of HPS content causing the increment of free volume in the matrix. The presence of free volume in the matrix also described in [9] about the free volume theory of plasticizer in the synthetic polymer system. This condition already been reported by previous study by using LDPE and crude palm oil in a blend [23]. Plus, [6] also mentioned that the presence of palm oil content increased the chain mobility of in the HDPE matrix and altered the mechanical behaviour of the blends.

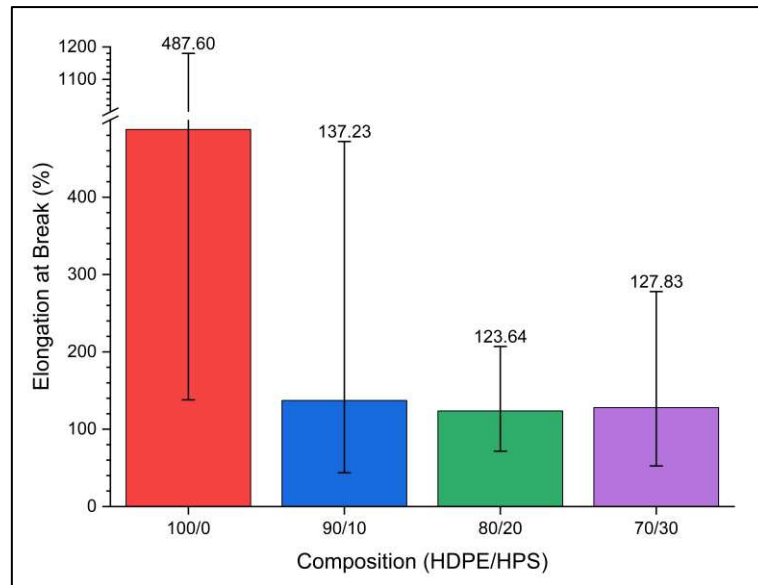


Fig. 7 Elongation at Break of HDPE/HPS Blends

Fig. 7 shows the elongation at break of the HDPE and HDPE/HPS blends, respectively. Based on the figure above, the elongation at break value portrayed a sudden 71.9% reduction after 10 wt.% HPS content was added to HDPE, which were from 487.60% to 137.23% elongation. However, the reduction value was maintained as the amount of HPS increased up to 30% HPS (specimen 70/30). This characteristic can be explained by the reduced ductility of HDPE after HPS addition, causing the specimen to undergo lower elongation before specimen's fracture. This result does not fully follow the previous studies by Ratnam et al. [6] since that the range of palm oil content used in this study was higher than previously. The previous studies only discovered the mechanical properties of HDPE with below than 10% palm oil content involved [6, 23] which may impact the tensile properties shown, possibly reflected a large gap of plasticization effect of palm oil content.

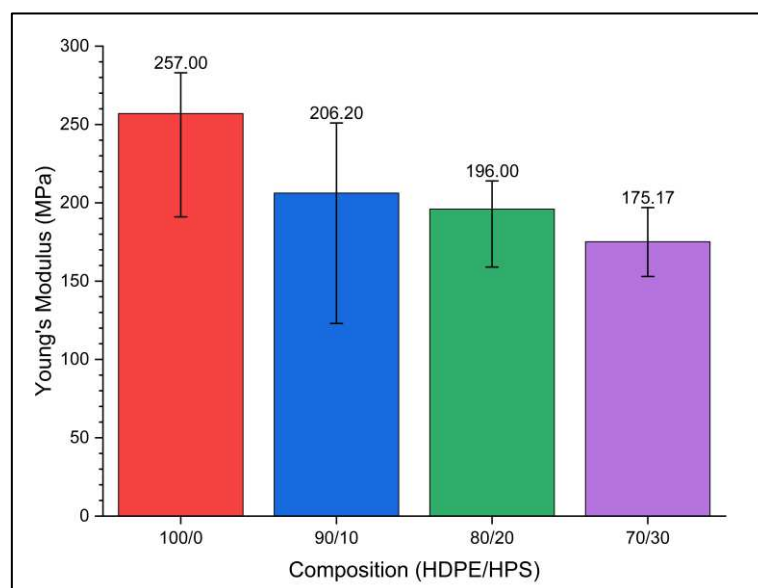


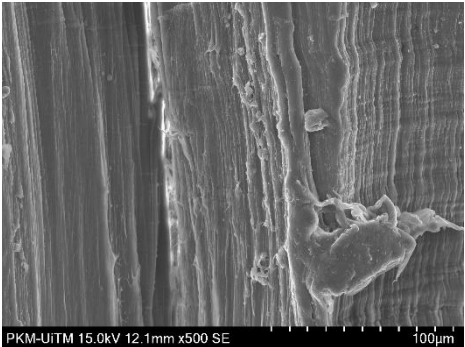
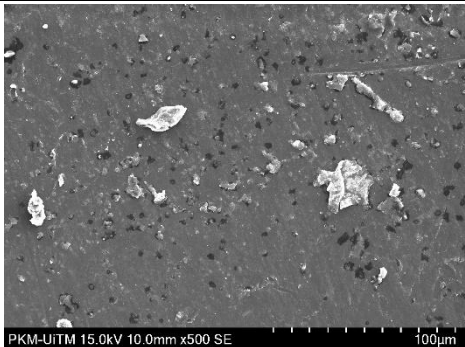
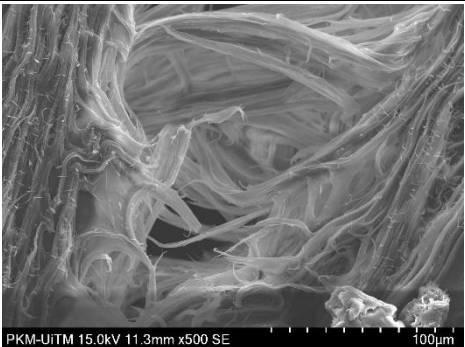
Fig. 8 Young's Modulus of HDPE/HPS Blends

Fig. 8 shows the Young's modulus (modulus of elasticity) of the HDPE/HPS blends. The result showed the decrement of Young's modulus from 257.00 MPa until it reaches 175.17 MPa as the 30wt.% of HPS added to HDPE. The reduction of the value was described as the decrement on stiffness of the HDPE as it plasticized with HPS. The reduction of the Young's modulus changes slowly from 19.8% to 23.7% and 31.8% as the amount of HPS increased from 10% to 20% and 30% respectively. The allocation of the plasticizer does lead to a decrease in stiffness of the blends due to enhancement of chain mobility after plasticization. This description has a consistent trend across different polymer systems such as plasticized PLA, plasticized poly (methyl methacrylate) and thermoplastic starch [24–26].

3.3 Scanning Electron Microscopy (SEM)

Table 2 shows the images of scanning electron microscopy (SEM) at 500x magnification for the two different specimen's category which is virgin HDPE (100/0), plasticized HDPE (80/20). Two different morphological cross-section conditions, which are before and after tensile testing on the gauge (tensile testing break) area are shown in the table below.

Table 2 SEM Images of Before and After Tensile Fracture Zone at 500x Magnification

Composition	Before Tensile Break	After Tensile Break
100/0		
	Image (a)	Image (b)
80/20		
	Image (c)	Image (d)

Images (a) and (c) shows the cross-section of 100/0 and 80/20 structure before tensile test. Image (a) shows compact structure with a minor rough surface of the HDPE which portrayed a strong matrix integrity and the image result can be supported by previous study by

Ratnam et al., 2006 [6]. Meanwhile, Image (c) shows a clustered of dark pores / void on the surface of the specimen 80/20. This effect was due to the plasticizer fusion which possibly leads to the partial phase separation and created free volume formation due to the weak intermolecular forces. The increment of the dark pores' intensity could be supported the reduction of the specimen stiffness in the Young's modulus analysis of the specimen neat HDPE (100/0) which is 257 MPa to 196 MPa after plasticization occur at the specimen 80/20. This situation helped the sample increased flexibility and easily to deform. Plus, the formation of a smoother structure (80/20) compared to the neat HDPE (100/0) which showed a good plasticization result. The plasticization effect of palm oil content towards polymer matrix description was referred to the previous study [27].

Image (b) and (d) shows the deformation pattern from the cross section of the specimens 100/0 and 80/20 after tensile break. Image (b) showed up with a consistent stretching pattern and minor deformation pattern during tensile break as followed the morphology image shown by Ratnam et al., 2006 [6]. Meanwhile, the plasticized HDPE (80/20) showed a unique stretching deformation. The deformation pattern in image (d) showed that how the HPS can give flexibility towards HDPE matrix. The effect of plasticization clearly recorded by the shape of the specimen's deformation after plasticization (80/20) less rigid compared to the specimen before plasticization (100/0) which can supported the reduction of the tensile strength value from 22.68 to 15.41 MPa as shown in Fig. 6. Evidently, presence of void / cavities in image (c) before tensile break become one of the causes of the ductile fracture that leads to a further failure at a lower elongation during tensile testing as shown in Fig. 7.

3.4 *Differential Scanning Calorimetry (DSC)*

Fig. 9 and Fig. 10 show the heating and cooling thermograms of HDPE/HPS blends with different HPS loadings. The thermal transitions observed the melting temperature (T_m),

crystallization temperature (T_c), and glass transition temperature (T_g) of the HDPE/HPS blends. The thermal transitions mentioned was tabulate in Table 3.

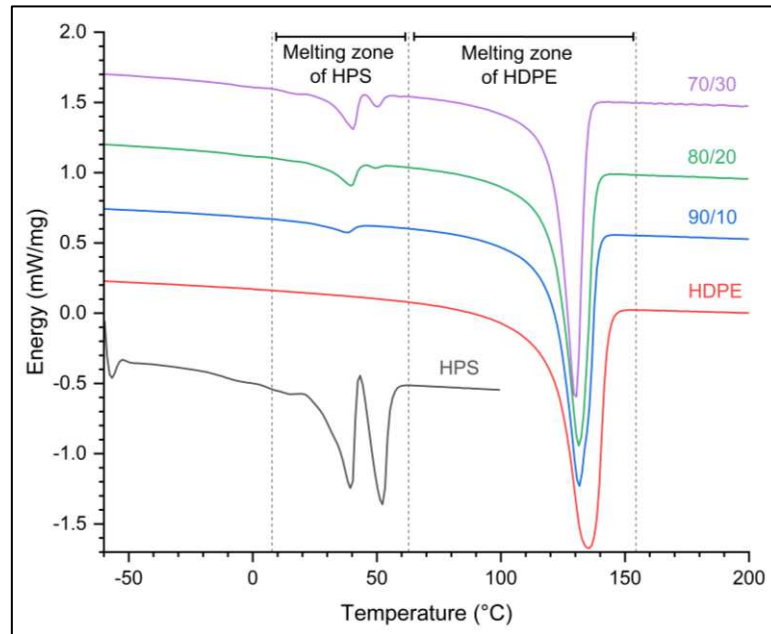


Fig. 9 Heating Curve Thermogram of HDPE/HPS Blends

Table 3 Thermal Properties of HDPE/HPS Blends

Material blends		HPS	HDPE	90/10	80/20	70/30
HPS	$T_{g, HPS}$ (°C)	-40	-	-	-	-
	$T_{m1, HPS}$ (°C)	39.6	-	38.3	39.7	40.4
	$T_{m2, HPS}$ (°C)	52.2	-	-	-	-
	$T_{cl, HPS}$ (°C)	24.7	-	28.3	30.8	32.8
	$T_{c2, HPS}$ (°C)	32.4	-	-	-	-
HDPE	$T_{m, HDPE}$ (°C)	-	135.2	131.5	131.4	130.0
	$T_c, HDPE$ (°C)	-	109.7	113.2	112.3	113.2
	ΔH_m (J/g)	-	201.7	167.2	169.7	147.1
	X_c (%)	-	68.7	63.4	72.4	71.7

Based on Table 3, the neat HDPE (100/0) exhibited a melting temperature ($T_{m, HDPE}$) of 135.2 °C, while the HPS loading gradually reduced the melting temperature of the 90/10, 80/20 and 70/30 blends to 131.5 °C, 131.4 °C and 130.0 °C, respectively. Based on the previous study of HDPE plasticized by crude palm oil, the small amount of crude palm oil (2-5%) does gave a small decrease in melting temperature [7]. The reduction in melting temperature possibly caused by the incorporation of HPS disrupted the regular structure of the HDPE molecular chains. The difference on the melting temperature change may be due to high amount of HPS plasticizer. The same pattern of melting temperature of this study and previous study can be concluded that the palm oil-based plasticizer can reduced the melting temperature of HDPE.

Plus, the melting peak of the HPS was gradually increased from 38.3, 39.7 and 40.4 °C as followed the specimen 90/10, 80/20, 70/30 respectively. It is suggested that the HDPE in the

blend hindered the melting temperature of the HPS which caused the HPS content in the specimen 90/10, 80/20, and 70/30 melted at higher temperature compared to the virgin HPS.

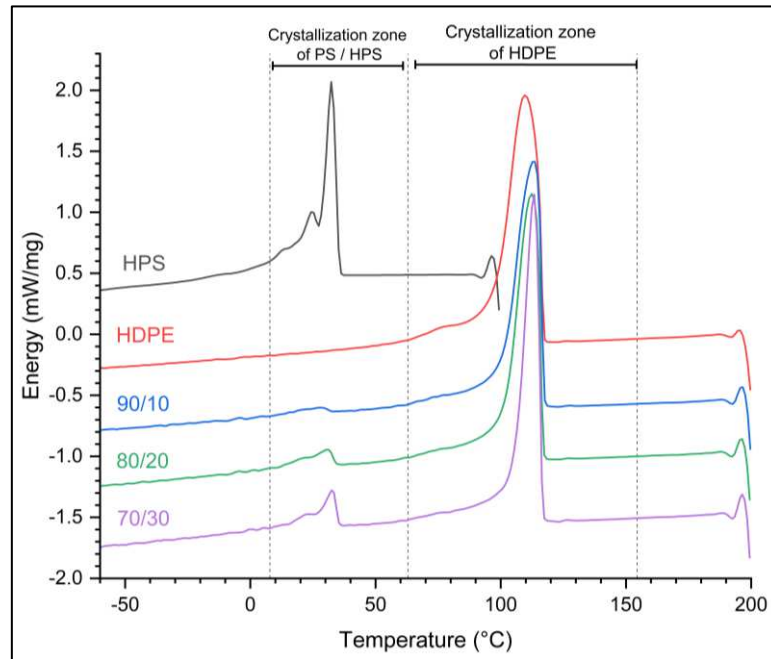


Fig. 10 Cooling Curve Thermogram of HDPE/HPS Blends

After that, the crystallization behaviour of the blends also as shown in Fig. 10. The neat HDPE crystallization peak appeared at 109.7 °C. By the addition of HPS plasticizer, the crystallization temperature of the HDPE ($T_{c, HDPE}$) increased up to 113.2 °C. Meanwhile, the blended specimens also exhibited the additional crystallization peak in the lower temperature zone between 28.3 °C and 32.8 °C. The area of the peak ($T_{cl, HPS}$) appeared larger as the HPS loading increased and the peak shifted from 28.3 °C, 30.8 °C and 32.8 °C as followed specimen 90/10, 80/20, 70/30 respectively.

This condition suggests that the presence of HPS plasticizer influenced the crystallization behaviour of HDPE while maintaining partially separated crystallization domains of each component in the blend. Plus, the specimen 70/30 clearly showed two separated crystallization peaks which represent HDPE and HPS component's crystallization peak. The specimen 80/20 condition start to show two endothermic peaks in the cooling curve in Fig. 10, which possible to explain that there is presence of immiscibility between HDPE and HPS when more than 20% HPS content was applied. As the HPS content increased to 30%, the endothermic peak clearly showed in two separate peaks. This condition has been explained in the study by Liu et al. which the presence of two separate endothermic peaks in the cooling curve confirmed the immiscibility of two materials in the blend [28].

The slight increment of HDPE crystallization temperature from 109.7 °C to approximately 113 °C also suggests that HPS influenced the crystallization process of HDPE during cooling process which possibly there are changes in HDPE's structure due to

plasticization. Plus, this condition also reflects the lack of interaction between HDPE and palm stearin content which disrupt the crystalline structure of the HDPE [29].

In addition, the melting enthalpy of the HDPE, ΔH_m tend to gradually reduce from 201.7 J/g to 147.1 J/g as the amount of HPS increased. This condition showed that presence HPS content assist in accelerating melting process of the HDPE. This condition can be supported by the pattern of the mixing torque curve in Fig. 5 which the final torque value reduced as the amount of HPS content increased in the blend. By lowering the melting enthalpy of the HDPE, the force applied towards material blending process will be reduced and reflected in the measurement of the torque value. However, the plasticization of HDPE using HPS does not showed any significant change in the degree of crystallinity of the HDPE. The degree of crystallinity of the HDPE was changed from 68.7% to 63.4%, 72.4% and 71.7% based on the HPS content 10-30wt% respectively. This pattern also showed in the previous study by Min Min et al. where, the application of crude palm oil as the plasticizer towards HDPE does not show major changes on the degree of crystallinity of HDPE [7].

3.5 Thermogravimetric Analysis (TGA)

The thermal stability and degradation behaviour of the HDPE/HPS blends were evaluated using the thermogravimetric analysis (TGA) as shown in Fig. 11 with various amounts of HPS loaded. The figure mentioned focusing on the onset of 5% weight degradation temperature ($T_{d5\%}$) which represents the temperature at which 5% of overall mass of the sample loss due to thermal exposure. The value of the $T_{d5\%}$ also can be referred to Table 4.

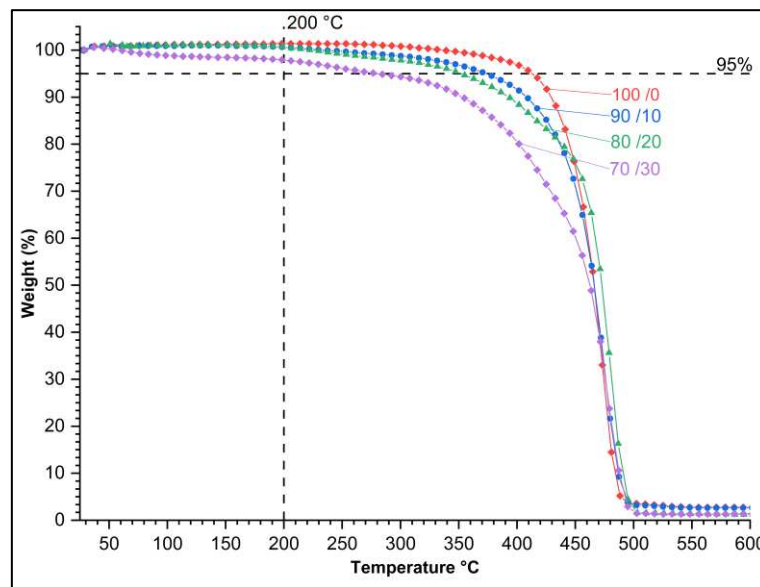


Fig. 11 TGA Curve of HDPE/HPS Blends

Table 4 5% Degradation Temperature of HDPE/HPS Blends

Sample Blend Type	Sample Name	$T_{d5\%}$ (°C)	% Change (%)
HDPE	100/0	413.0	-
	90/10	373.4	9.6
HDPE/HPS	80/20	351.9	14.8
	70/30	280.6	32.1

The thermogram in Fig. 11 showed a clear trend where the addition of hydroxylated palm stearin (HPS) reduces the thermal stability of the HDPE matrix when the temperature above the processing limit temperature (200 °C). The $T_{d5\%}$ of the HDPE which is 413.0 °C decreased up to 32.1% as the HPS applied up to 30 wt.%. This significant drop, particularly at higher HPS content, is attributed to the lower thermal stability of the bio-based plasticizer. The thermal profile of the plasticizer can influence the thermal behaviour of the plasticized polymer by lowering the thermal resistance and initiates the degradation process of the specimen at a lower temperature [30, 31]. It was stated that possibility the presence of plasticizer interacted with the synthetic polymer and enhance chain mobility which resulted in lowering thermal resistance. As the amount of HPS increased, the $T_{d5\%}$ decreased which reflected the decrement in thermal stability [32].

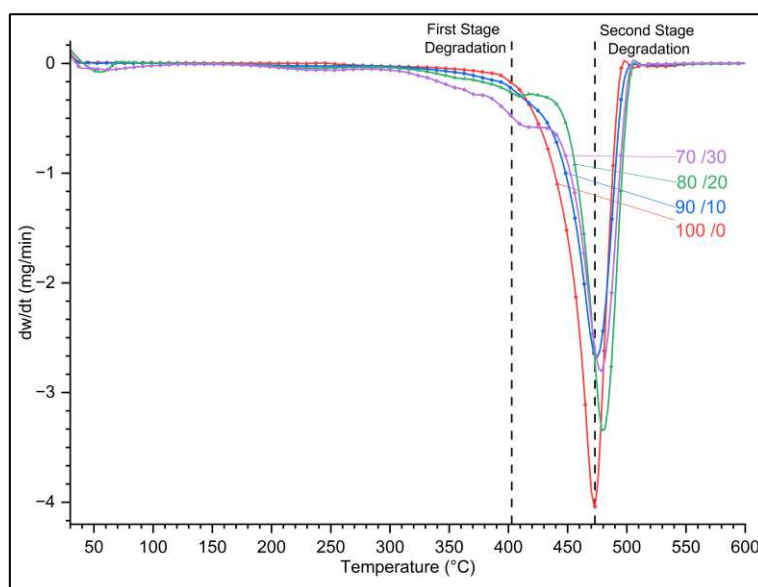


Fig. 12 DTG Curve of HDPE/HPS Blends

After that, the thermogram in Fig. 12 shows a single-step degradation peak for the blends 90/10 and 80/20. However, the specimen with 30% HPS (70/30) almost formed 2-stage degradation peak. This degradation peak also can be referred to the derivative thermogravimetry (DTG) curve in Fig. 12. This difference curves can be explained by the different miscibility properties of the blends mentioned. Previous study had explained that a good miscible polymer blend formed a single-step degradation peak as the heat applied to the specimen [32, 33]. As the amount of the HPS plasticizer increased, the degradation peak

gradually to change from single-step peak into two-step peak. The condition shows that the plasticizer amount influenced the thermal stability of the blends, where the possibility of the excess plasticizer gave the formation a two-step degradation instead of single-step degradation. As the amount of HPS increased, the thermal stability of the HDPE/HPS blends decreased.

3.6 Fourier Transform Infrared Spectroscopy (FTIR)

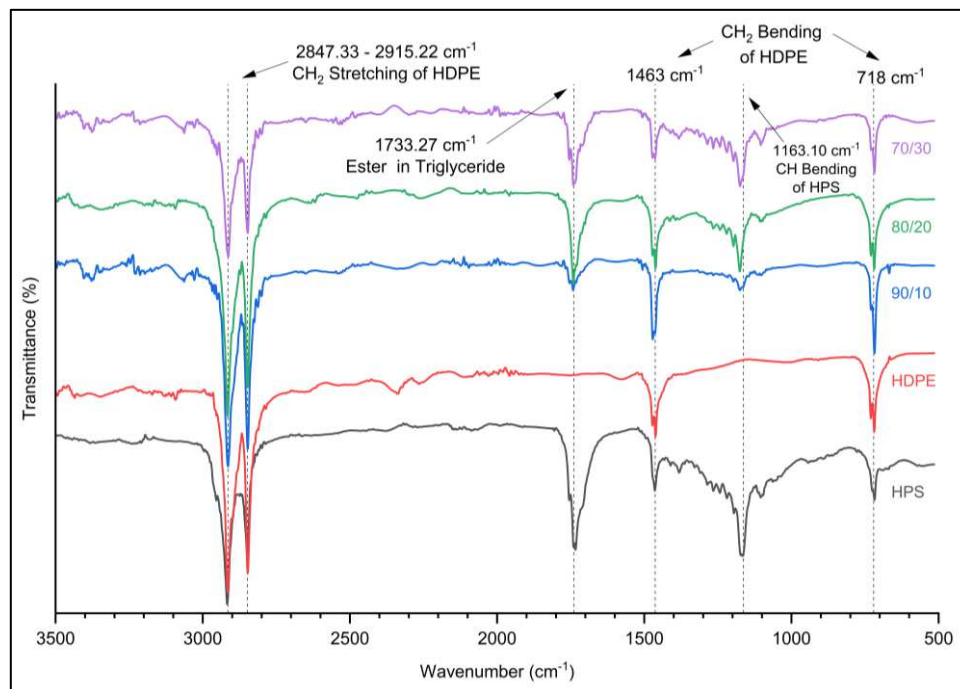


Fig. 13 FTIR Spectra of HDPE/HPS Blends

Fig. 13 shows the FTIR spectra neat HDPE, HPS plasticizer and all blend specimen (90/10, 80/20, and 70/30). In Fig. 13, the HDPE peak showed 2 main peaks represented the CH₂ bending which is 718 cm⁻¹, 1461.93 cm⁻¹. The other peaks appeared which is 2847.33 cm⁻¹ and 2915.22 cm⁻¹ represented CH₂ stretching. These peaks were comparable to the HDPE shown by Gulmin et al. and Charles et al. The peaks mentioned was 720 cm⁻¹ and 1463 cm⁻¹ which represents CH₂ bending of HDPE and 2 peaks between 2848 – 2915 cm⁻¹ which represents CH₂ stretching of HDPE chain [34, 35].

After that, the first main peak for the HPS plasticizer was 1733.27 cm⁻¹ which represents ester functional group (C=O) in triglyceride structure of the palm stearin [36]. The second peak was 1163.10 cm⁻¹ which represents C-H bending of the palm stearin structure [37]. Throughout the hydroxylation process of the HPS, some small peaks tend to be appeared in the range of 840 – 830 cm⁻¹ which represented the oxirane ring (C-O-C) from the epoxidation process [38] and the appearance of peak at 1381 cm⁻¹ which represented O-H bond of the hydroxylated palm stearin [36].

There were two small peak shifting happened for the all blends which C=O ester peak of HPS shifted from 1733.27 cm^{-1} to 1740.95 cm^{-1} and the second one was CH peak of HPS shifted from 1163.10 cm^{-1} to 1175 cm^{-1} . This condition possibly caused by the weak intermolecular forces between of the HPS that leads to chain mobility of HDPE increased after plasticization process without any new interaction peak formed in the spectrum. After blending process, the spectrum of blends 90/10, 80/20 and 70/30 retained the characteristic peaks of the spectra HDPE and HPS where there are no major peak loss / peak shifting after plasticization process occurred. So, this condition indicates that the HPS only play a role as a non-reactive plasticizer since there is no presence of new absorption peak [39]. This statement can be supported by the mixing torque curve in Fig. 5 which showed there are no post-reaction after plasticization.

Conclusion

This study successfully evaluated the plasticization behaviour of the hydroxylated palm stearin (HPS) towards the high-density polyethylene (HDPE). HPS can serve as an effective, bio-based plasticizer for HDPE with loading of 10-30 wt%, offering a viable alternative for synthetic additives. As seen by the significant drop in mixing torque, the addition of HPS successfully decreased the melt viscosity of HDPE, proving that it can increase processability and polymer chain mobility. Mechanical characterization revealed that increasing HPS concentration progressively reduced the tensile strength of HDPE up to 46.5% reduction as the HPS amount increased from 10-30 wt% due to weakened intermolecular forces. Plus, the morphology observation also supported this mechanism through the presence of few small voids' formation after plasticization due to plasticizer fusion which possibly leads to the free volume formation in the surface structure. Thermal analysis also showed that HPS increased HDPE's crystallization temperature while slightly lowering its melting temperature. The major reduction of the melting enthalpy of HDPE from 201.7 J/g to 147.1 J/g as the amount of HPS increased assisted in accelerating the melting process of the blends. As seen by the drop in the 5% degradation temperature ($T_{d5\%}$) at 30 wt% HPS, the thermal degradation behaviour demonstrated that increasing HPS loading progressively reduced the thermal stability of the blends which is good properties from accelerated degradation for the biohybrid plastic. Lastly, the small shift of the C=O ester structure and C-H of HPS possibly related to weak intermolecular forces that leads to the increment of HDPE's chain mobility without any new interaction peak formed in the spectrum. To further improve blend homogeneity and increase the permitted amount of bio-based plasticiser without sacrificing material performance, future research should concentrate on enhancing the compatibility between HPS and HDPE through reactive compatibilization or chemical modification.

Author Contributions

Research conceptual idea: N.A. Ramlee & I. Subuki. Sample preparation and analysis: R. M. Shariff & A. N. Mazni. First draft writing: R. M. Shariff. Mechanical analysis: A.N. Mazni. Final draft writing and approval: N.A. Ramlee & R.M. Shariff.

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