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THE EFFECT OF BLENDS FORMULATION ON PROPERTIES OF THE RECYCLED HIGH DENSITY POLYETHYLENE (rHDPE) AND RECYCLED POLYPROPYLENE (rPP) FROM INJECTION MOLDING SCRAP

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Abstract: *The effect of blends formulation on properties of the rHDPE and rPP on the mechanical, physical and thermal properties were studied. Different ratio of rHDPE/rPP (0/100 wt.%, 30/70 wt.%, 50/50 wt.%, 70/30 wt.%, 100/0 wt.%) are used to investigate the effects of materials phases towards the resulted mechanical, thermal and physical properties of rHDPE/rPP blends. Later, the fractured surface morphology of the selected rHDPE/rPP blend samples are analysed through Scanning Electron Microscopy (SEM) observation. In addition, the melting temperatures (T_m) of rHDPE/rPP samples are measured by Differential Scanning Calorimeter (DSC) technique for miscibility evaluation through thermal route. Overall, from this study it was found that rHDPE/rPP blend with higher strength and thermal performances is proposed for substituting the Delrin material for machining application. It was found that the 70/30 wt.% of rHDPE/rPP was significantly possessed outstanding mechanical and physical characteristic in terms of their tensile, elongation at break and the hardness extraordinary improvement at about 59.8% in their tensile strength, 473% of elongation at break behaviour, as well as better thermal properties improvement. The 70/30 of rHDPE/rPP blend has yielded 2.3% of the hardness improvements. It was also found that there are two T_m peaks appeared in the total heat flow curve for all formulated blends which indicates the immiscibility nature of produced blends. The range of T_m was found gradually decreased when the portion of rHDPE phase is increased up to 100 wt.%.*

Keywords: *r-HDPE/r-PP recycled blends; injection molding scrap; sustainable; miscibility.*

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1. Introduction

Massive of plastic waste is one of the global environmental issue and becoming tough challenge for most of the developing country (Gu and Ozbakkaloglu 2016, Iwata 2015, Singh et al. 2016). The amount of plastic wastes disposal are getting increasing due to increase of development and industrial activities, population growth, socio-economic and urban lifestyle. Less awareness in our society pertaining to the issue that contributes to the plastic waste disposal problem is getting alarming and troublesome.

Waste management is becoming a crucial issue in industrialized and developing country. The waste management without proper mechanism will surely affecting not only the environment but, to the society and economy of the country (Al-Salem et al. 2009, Salih et al. 2013). From the perspective of this study, the recycling of recyclable plastic waste materials which generated from an injection molding process has been the main interest of this study.

People were arguing about the environmental impact of plastics goes outwardly based on facts and logic and based on values and emotion. Aside from the view of plastic industry people, they are proud of the capabilities of their products and of the unique qualities of plastic materials as compared than other engineering materials (Tolinski, 2012). On the extreme other side, the environmental activists are particularly sensitive to the waste resources and their impact damaged towards the ecological and earth system that are mainly affecting the future generation (Verdolotti et al. 2014).

Recycling the polymer is a noble approach to reduce the environmental pollution issues that causes by the polymeric waste generation from everyday utilization of polymer materials such as household, parts and construction (Bernardo, et al. 2016). The recycling effort of polymeric waste could help to conserve the natural resources as the main backbone materials for polymers are the hydrocarbon from oil and natural gaseous.

This study is crucial to find the effect of blend composition of recycled polymeric materials blends formulation on mechanical, physical and thermal properties.

2. MATERIALS AND METHODOLOGY

The main raw material, rPP and rHDPE scrapped from injection moulding (IM) operation from IKTBN Pagoh, Malaysia has been utilized in this study. Five blends formulation of rHDPE/rPP materials were prepared accordingly; both materials are supplied by Parking Plastic Sdn. Bhd. The typical properties of PP and HDPE that are used to prepare the blends are listed in the following Table 1.1.

Tabe 1.1: Typical Properties of HDPE and PP (Parking Plastic SDN. BHD.)

ATTRIBUTES/MATERIALS	PP	HDPE
Maximum operating temperature, ^{0}C	90	82
Water absorption, %	0.03	0.2
Tensile strength, MPa	52.0	27.6
Flexural Modulus, Mpa	1200	1380
Notched Izod, kJ/m	0.1	0.2
Density, g/cm^3	0.92	0.90
Melting point, ^{0}C	160	180

1.2 Preparation of rHDPE and rPP Formulation Blend

rPP was mixed into rHDPE with different weight ratios which varied from 30% to 70 % were prepared based on per kilogram weight. All of the prepared mixtures are loaded into the crusher machine and the materials were crushed into fine particles. About five(5) different weight percentage (wt %) formulation of rHDPE/rPP blends preparation as given in the Table 1.2. There are no additives added into these blends except as available colourant pigments from the injection moulding process.

Table 1.2: The Composition of PP and HDPE

POLYMER	A1	A2	A3	A4	A5
HDPE, wt %	100	70	50	30	-
PP, wt %	-	30	50	70	100

1.3 Mechanical and Physical Testing of Blend Formulation of rHDPE/rPP

There are two types of mechanical testing that has been carried out in this research, which is tensile testing for mechanical properties determination. While shore-D hardness for physical properties evaluation. All of the testing that were performed in this research is in accordance to the ASTM standard as tabulated in the following Table 1.3.

Table 1.3: ASTM Standard for Mechanical and Physical Testing

TESTING	STANDARD
Tensile	ASTM D638 Type I
Hardness	Shore D hardness, D Type

1.3.1 Tensile Testing Using Universal Testing Machine

The samples were prepared by cutting the rectangular compressed samples into the dumbbell shape. The samples then were prepared on 20kN universal testing machine (UTM:Shimadzu, AGS X) to determine the tensile strength and elongation at break of each blending ratios. A crosshead speed was set into 20mm / min.

1.3.2 Shore-D Hardness Testing

Shore-D Hardness tester is the standard model for measuring the soft hardness (Shore-D). The device is consisted of a measurement unit with sensor and 360° anti-glare dial with an accuracy of 0.50 hardness units. To test the hardness of the rHDPE and rPP blends samples at five (5) different points of each sample were measured to get the data of each tested samples.

1.3.3 Differential Scanning Calorimetry (DSC)

The differential scanning calorimeter (DSC) machine set-up model Jade from Perkin Elmer, which has been utilized in the analysis. The sample which about 5-15 mg was stored in an inert metal pans for the thermal analysis. The pans will then be placed inside the furnace and the temperature was raised up to 200 °C. The measurement was recorded between 25 °C and 200 °C at a heating rate of 10 °C/min. From this thermal analysis, the miscibility of the blend samples could be inspected from the detection of T_m peaks produced in the temperature scan.

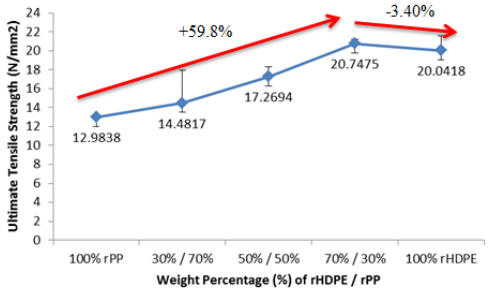
1.3.4 Scanning Electron Microscope (SEM)

The morphological feature of fractured sample was obtained by scanning the sample with a strong electron beam. By using SEM, the surface failure mode can be observed and was further analysed. Furthermore, by using SEM, the distribution of rHDPE/rPP blend phases could be observed by providing the image of surface features. Macroscopic analysis was performed by visual inspection of the fractured surfaces using a stereoscopic microscope. About 5 mm sections of all the fragments were subjected to SEM for microscopic analysis to verify their fracture morphology.

3. Result and Discussion

Figure 1.1 shows the ultimate tensile strength result of various weight percentages ratio of hybrid materials of rHDPE and rPP. It was obviously observed that the ultimate tensile strength are found tremendously increased with the increase in weight percentages of rHDPE addition, until an optimum point at 70 wt. % of rHDPE and 30 wt. % of rPP, before it was significantly dropped around 3.40 % after further addition of rHDPE at higher percentages content up to without any presence of rPP. The largest increase of ultimate tensile strength is about 59.8 % yielded at 70 / 30 wt. % of rHDPE/rPP blend. It was found that the tensile strength has increased with only about 30% amount of rPP and significantly dropped without the existence of rPP phase. It was likely due to the stiffer polymer grades present in the recycled plastic dominated by rHDPE phase. This is due to the presence of mixed type of polymer and chemical impurities in the rHDPE and rPP, which enhanced the interface bonding between both phases. In addition, the tensile strength of the blending materials has increased with the decreasing content of rPP in the produced samples. However, this finding was contrary with Atikah et al. (2014) who found that the tensile strength of rHDPE/rPP blends are increased with the rPP contents. It can be seen that all blends have significantly increased in their tensile strength with the increasing content of rHDPE. This may due to the presence of rPP which may interrupt the crystalline structure homogeneously of rHDPE due to rPP amorphous characteristic that happened when the methyl group in rPP chain exhibit no preferred orientation and become atactic (Montes et al. 1998). Increase in rPP content has obstructed the ordered arrangement of rHDPE and decreased their crystallinity, which in turn caused increasing in tensile strength value. Thus, the higher the rPP content in the blend,the lower the ultimate tensile strength values of rHDPE/rPP blend performance.

Figure 1.1: The Effect of Different Weight Percentages Ratio Between rHDPE and rPP Composition into the Ultimate Tensile Strength of rHDPE/rPP Hybrid Blend



The following Figure 1.2 depicts the modulus of elasticity results of rHDPE and rPP based polymer blend materials. The plotted graph shows the effects of various weight percentages addition of rHDPE into the rPP. The addition of rHDPE has increased in the Young's modulus up to 19.5 % enhancement before decreased afterwards about 18.1 % with the increasing of rHDPE portion. The trend was almost similar to the ultimate tensile strength plots attributes. The highest Young's modulus was found at about 8.21 N/mm^2 which belongs to the composition of 50 wt. % of rHDPE and 50 wt. % of rPP. The lowest Young's modulus was detected from the sample with the composition of 100 wt. % of rHDPE which is 6.72 N/mm^2. There are almost 18 - 20 % of reductions as compared to composition without existence of rHDPE or rPP. The properties dropped were might due to the improper dispersion of rPP into rHDPE at higher concentration which closely related to the contaminated material effect. Figure 1.5 (b) depicts the micrograph of 100 wt. % of rHDPE sample. The fracture morphology observation via SEM has clearly shown that some contaminations are detected due to bad dispersion of rHDPE. These contaminations indicate unmelted rHDPE phase at their matrix counterparts. These contaminations will act as a source of crack concentration and crack initiator point that lead into lowering the Young's modulus of produced blend samples (Garg et al. 2011).

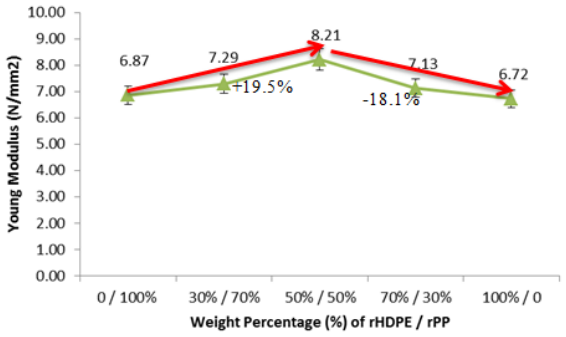


Figure 1.2: The Effect of Different Weight Percentages of rHDPE and rPP Composition Ratio into the Modulus of Elasticity of rHDPE/rPP Blends

Hence, the addition of rHDPE gives significant increment to the elongation at break of rHDPE/rPP blends as shown in the following Figure 1.3. At different blend ratios of rHDPE/rPP, the elongation at break has experienced decreased pattern. This is due to the presence of additives in the rPP that normally could give rigid behaviour into the recycled based thermoplastic. This may due to the factor of having good interfacial interaction between the hybrid reinforcement. Therefore the sample is getting stiffer and brittle as represented by the lower percentages of elongation at break. However, at 70 wt. % of rHDPE addition, the produced sample shows the highest percentages of elongation at break, which is about 473 % enhancement. This is because when the rHDPE is further added, the viscosity of blended materials had increased significantly. Thus resulting a serious agglomeration of rHDPE macromolecular chain which later preventing proper interaction between these blend phases of rHDPE and rPP.

The presence of additives from recycled materials has improved the adhesion at the interfaces between rHDPE and rPP phases which responsible in increasing the stiffness of blends. However, it also caused a notable reduction in the elongation at break plots. Additionally, the addition of

additives has increased the interlocking condition between two phases, thus increasing the interfacial adhesion between phases in polymer blend state. This would lead to low mobility of the chains at rHDPE/rPP blend interface and has significantly reduced the elongation at break at lower percentage of rHDPE or rPP phase.

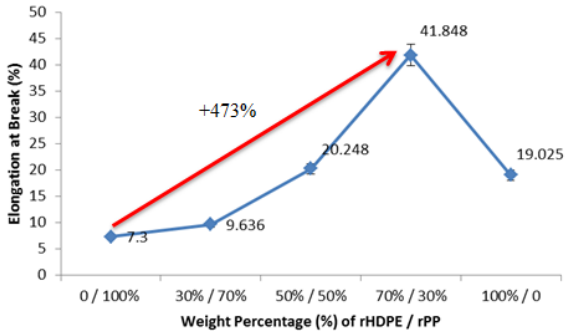
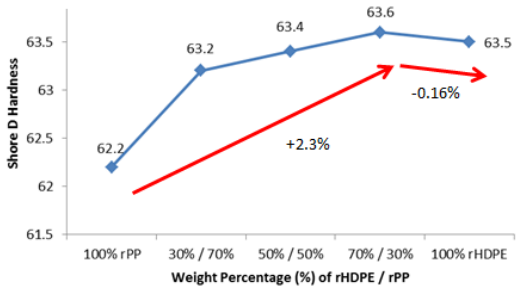


Figure 1.3: The Effect of Different Weight Percentages of rHDPE and rPP Composition into the Elongation at Break of rHDPE/rPP Blends

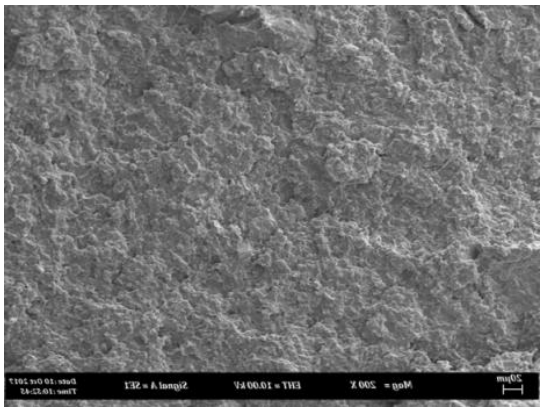
The variation in Shore-D hardness values of rHDPE and rPP is presented in the following Figure 1.4 by using Shore-D hardness tester. Although the fact that PP is stronger and more rigid than HDPE (Smith and Hashemi, 2006 and Shank et al. 2007) but the Shore-D results of this recycled material batches shows oppositely from these facts since PP has higher hardness value than the other polymeric types. Furthermore, it was noticed that the Shore-D values for rHDPE / rPP blends has increased as the weight ratio of rHDPE increased about 2.3 % up until high value of rHDPE portion into rPP phases, but the value was drop into 0.16 % without any existence of rPP. From this plot, it has been noticed that the highest value recorded at 63.6 Shore-D hardness values for 70/30 rHDPE/rPP. The rPP without the existence of rHDPE would give the lowest value of Shore-D hardness. This result was opposite from Salih et al. (2013) who stated that Shore-D values for HDPE/PP and LDPE/PP blends has increased as the weight ratio of PP increased. This is due to the nature of PP which is stiffer than any one of PE types.

Figure 1.4: The Effects of Weight Percentage of rHDPE and rPP Into the Hardness of the rHDPE/rPP Recycled Based Polymeric Blends

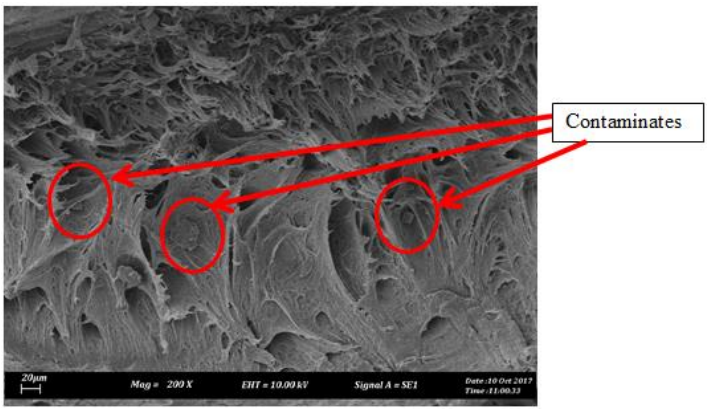


Scanning Electron Microscopy (SEM)

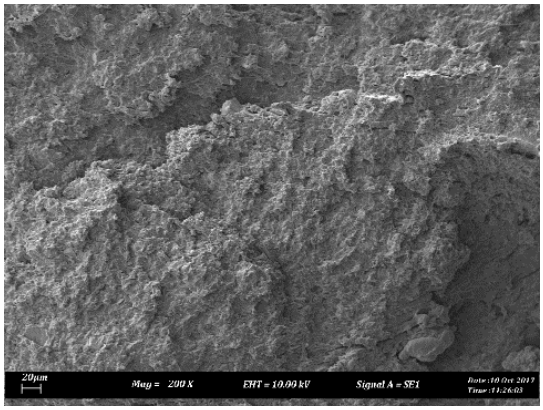
Micrograph of the following Figure 1.5 (a), (b), (c), (d) and (e) show the fractured surface of rHDPE/rPP blend samples at different composition of rHDPE and rPP phases which are 100 wt. % of rPP, 100 wt. % of rHDPE, 30 wt. % of rHDPE and 70 wt. % of rPP, 50 wt. % of rHDPE and 50 wt. % rPP, 70 wt. % of rHDPE and 30 wt. % of rPP. The following Figure 5.0 (a) and (b) present the homogeneous surface of rPP and rHDPE which exhibit rHDPE ductile tearing mode and rPP brittle tearing mode. The SEM micrograph of rHDPE/rPP blend at wt. % of 30/70 blend ratio has indicated rougher surface as shown in the following Figure 1.5 (c). The 50/50 wt. % of rHDPE/rPP micrograph has clearly indicates that the rHDPE and rPP are incompatible phase separation towards each other. For example, if rHDPE induces a change transformation in phase continuity, changing from a dispersed rPP and rHDPE phase as shown in the following Figure 1.5 (a) and (b) to a more continuous phase as seen in the following Figure 1.5 (c) and (d) are portrayed by an increase in mechanical properties of the respective blend are seen.



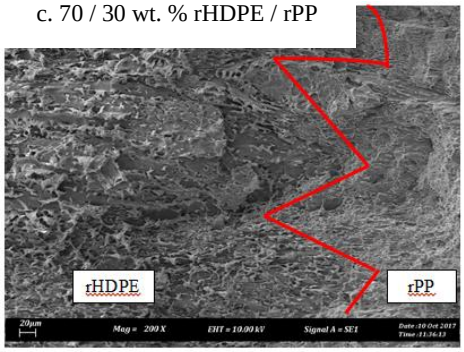
a. 100 wt. % rPP



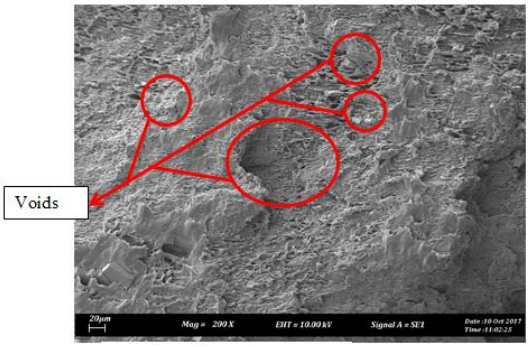
b. 100 wt. % rHDPE



c. 70 / 30 wt. % rHDPE / rPP



d. 50 / 50 wt. % rHDPE / rPP



e. 30 / 70 wt. % rHDPE / rPP

Figure 1.5: Fracture Surface of rHDPE/rPP Blend Samples with Different Composition of rHDPE and rPP Which are a) 100 wt. % of rPP, b) 100 wt. % of rHDPE, c) 70 wt. % of rHDPE and 30 wt. % of rPP, d) 50 wt. % of rHDPE and 50 wt. % of rPP, e) 30 wt. % of rHDPE and 70 wt. % of rPP

Heat Flow Analysis by Differential Scanning Calorimeter (DSC)

Thermal analysis on rHDPE and rPP blends shows that there are two peaks of melting point in the total heat flow curve for all blends ratios as shown in the following thermogram as available in the Figure 1.6. These mean that all the blends are immiscible. This result had supported by the observation done by Madi (2013). From the thermogram, it was noticed that 100 wt. % of rHDPE and rPP also have two peaks which confirmed that the recycled materials itself has already contained both materials with unknown composition. Surprisingly, the DSC thermogram also shows two peaks of virgin HDPE that come from recycled raw material as shown in the following Figure 1.7.

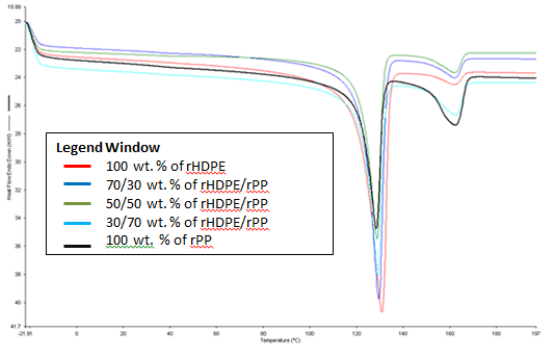


Figure 1.6: Overlaid of DSC Thermograms for rHDPE and rPP Blend Polymers

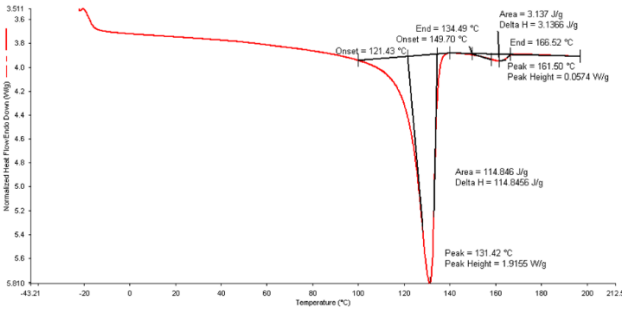


Figure 1.7: DSC Thermogram of Recycled Raw Material of rHDPE

Interesting characteristics can be obtained from these results. For blends of rHDPE/rPP, the incompatibility between rHDPE and rPP phase seems to be preserved. Two peaks on melting were appeared in the total heat flow curve: one for rHDPE and another for rPP. It is well known that rHDPE/rPP is a heterogeneous two-phase system, the components of which crystallize separately into discrete phases (Tao et al., 2007, Montes et al. 1998). It seems that, there is a limited miscibility of rPP in rHDPE especially at lower contents of rPP as shown in the following Figure 1.6. It can be seen that the T_m peak of rHDPE in all blend compositions shifts to higher temperatures. In addition, the T_m of rPP shifts to lower temperatures in relation to its composition in the blends, which indicates a reduction in the perfection of the formed crystallites. This result supported by Aumnate et al. (2016). This might be explained by less regular molecular structure and higher polydispersity index of the rHDPE resulted from oxidation and chain scission undergone during materials reprocessing. However, in most blends a depression in the T_m of recycled rHDPE is observed with increasing rPP content indicating that the degree of crystallinity is reduced by the blending. It was reported by Madi (2013) for the case of incompatible blends, the melting point decreases since the noncrystallisable component retard the crystal growth, which leads to imperfect crystals.

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4. CONCLUSION

Combining the rHDPE/rPP into hybrid blend, able to improve the properties of rHDPE and rPP phase. The ultimate tensile strength (UTS) and elongation at break of sample 70/30 wt. % of rHDPE / rPP are the highest among others sample, which is 20.7475 N/mm^2 and 41.84 % respectively. The Young's modulus and flexural strength of sample 50/50 wt. % of rHDPE/rPP are the highest among others sample, which 8.21 N/mm^2 and 19.68N/mm^2 respectively**.** Shore-D hardness for the material increase with the increasing of rHDPE contents. The most hardness material is for the sample of 70/30 wt. % of rHDPE/rPP which is 63.6. The hardness differentiation among samples is not too much only in the range 2.2 %. Rough material surfaces and possible nano-bridging were contributed to the betterment of the interfacial bonding between the blended materials that resulting better mechanical properties of produced hybrid recycled polymers. This is because of well dispersion of rHDPE and rPP that will lead into more effective load transfer. Heat flow analysis by Differential Scanning Calorimeter (DSC) for all recycled materials including virgin materials have two melting temperature (Tm) which is found incompatible each other. The range of Tm was found decreased gradually when the loading of rHDPE is increased up until 100 wt. % of recycled raw material of recycled HDPE.

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