

## THE EFFECTS OF r-HDPE/r-PP FORMULATION RATIO INTO MECHANICAL, THERMAL AND MORPHOLOGICAL BEHAVIOR OF r-HDPE/r-PP POLYMERIC BLENDS

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**Abstract** This study has reported the effects of different formulation ratio between recycled high density polyethylene (r-HDPE) and recycled polypropylene (r-PP) into the resulted mechanical, thermal and morphological properties of r-HDPE/r-PP polymeric blends. About five (5) different formulation ratio of r-HDPE/r-PP have been prepared and tested. The best combination ratio between r-HDPE and r-PP was determined in this work. It was found that the 70/30 wt.% of r-HDPE/r-PP blend possessed an outstanding mechanical and physical strength. About 59.80% and 2.30% of positive improvement in comparison to 0/100 wt.% of r-HDPE/r-PP was achieved for both of tensile strength and hardness, respectively. Interestingly, for 70/30 wt.% of r-HDPE/r-PP blend had also experienced major increased in their elongation at break up to 473%. The fracture morphological behavior of the tested samples that were observed via SEM observation, had established the interaction between the structure and properties of produced r-HDPE/r-PP blends, especially on the miscibility state between the r-HDPE and r-PP phases. Thermal evaluation by using the DSC had confirmed the partial miscibility state due to dominant peak shifting at 120 - 140°C and obvious melting peak reduction pattern. Overall, from this study, it was found that the blending between r-HDPE and r-PP into r-HDPE/r-PP blends are feasible to improve the properties of primary phase.

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

### 1. Introduction

Delrin or polyoxymethylene (POM) has been utilized as machining block for hands-on laboratory training for drilling, milling and grinding operation. Utilization of plastic based machining block are acquired to enhance student's confidence level prior real machining practice by using the metal block. POM, also known as polyacetal is an engineering thermoplastic used in precision parts requiring high stiffness, low friction and excellent dimensional stability. Delrin has been selected

considering its versatility and high strength behavior which able to sustain higher load due to machining process. Practical teaching and learning for machining by using Delrin as the machining block are really efficient for built-up confidence among students and are successfully reducing the waste generated if using metal block due to machining error. Even though the cost of polyacetal or polyformaldehyde are much cheaper than the cost of metal block, however, at the long run it was still not sustainable as for skill development, too much practice and trial and error approach need to be involved in effective teaching for machining. This has been generated higher total cost for raw materials and waste management for after-

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used Delrin machining block. This situation need to be solved by providing another low cost alternative to replace Delrin as one option of machining block for skills training purposes.

IKTBN Pagoh as one of special higher learning institution for youth technical development in Malaysia, has providing skill training in various highly-demanded industrial field. Other than industrial machining, this institute also providing special training in an injection molding (IM) operation and process design by using IM. Mass utilization of virgin polymer during the training period has producing massive waste generation due to purging, processing trial and error stage. This situation has created disposal and waste management problem that require innovative approach for an effective internal solution. Among the generated waste produced in-house from the operation of IM are high-density polyethylene (HDPE) and polypropylene (PP). The environmentalist are particularly sensitive towards the waste resources and their damaged to the ecological and earth system, which mainly effecting future generation [1]. Plastic waste management is becoming global industrialize issue which effecting much to the society and country economy [2, 3]. It becoming really tough challenge especially for most of the developing countries [4, 5, 6].

Dealing with this matter, in this study the recycling approach for both type of generated waste has been performed. Recycling the polymer could solve major disposal issue of daily plastic utilization from house-holds and other potential sources. This would conserve the environment and hydrocarbon natural source at the long run [7]. Considering the high strength attribute of HDPE and high temperature resistance of PP polymer, thus, both recycled high-density polyethylene (r-HDPE) and recycled polypropylene (r-PP) were blended together at different formulation ratios. The effects of formulation ratio into the resulted mechanical, physical, thermal and morphological properties of r-HDPE/r-PP blends were evaluated. The findings has been very crucial to provide an alternative raw materials for machining block made from

polymer materials, to replace the utilization of Delrin and previous metal based machining block. Interestingly, in this work, polymeric blend from the IM waste source has been utilized for r-HDPE/r-PP based machining block. In this work, the characterization results of r-HDPE/r-PP blends mechanical, thermal, physical and morphological properties were reported.

## 2. Materials and Methods

### 2.1 Materials

r-HDPE and r-PP scrapped from an injection molding operation at IKTBN Pagoh, Malaysia has been utilized as main raw materials in this study. Virgin HDPE and PP prior of IM processing stage has been supplied by the Parking Plastic Sdn. Bhd. Both r-HDPE and r-PP scrapped has been collected in-house from one-cycle of IM operation. The following Table 1, has been summarized the typical properties of as-supplied virgin HDPE and PP based on the supplier data set. About five (5) different formulation of r-HDPE/r-PP blends has been developed in this work (0/100 wt.%; 30/70 wt.%; 50/50 wt.%; 70/30 wt.% and 100/0 wt.%). For r-HDPE/r-PP blends preparation, the different ratio was prepared and weighted based on per kilogram weight basis. All of weighed r-HDPE and r-PP based on their specific ratio was loaded into the mechanical crusher for mixing and refining into fine particles mixture. No other additives has been mixed to the mixture, except the colorant pigment that has been added during the IM process into PP and HDPE. Later, after the mixing and crushing processes, the r-HDPE/r-PP mixture was hot pressed into specific dimension (20 cm x 20 cm x 2 mm) in accordance to their testing requirement, by using the hot press machine (brand GoTech) at 200°C using pressing pressure at 150 mBar. Produced r-HDPE/r-PP blend samples were conditioned at 80°C within 24 hrs. in a drying oven before performing the subsequent testing procedure. The Figure 1, summarized the overall flow of this study, started on material preparation and ended with results analysis.

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**Table 1.** Typical properties of virgin HDPE and PP (Parking Plastic Sdn. Bhd.).

Attributes / Materials	PP	HDPE
Maximum operating temperature, (°C)	90	82
Water absorption, (%)	0.03	0.20
Tensile strength, (MPa)	52.0	27.6
Flexural Modulus, (MPa)	1200	1380
Notched Izod Impact Strength, (kJ/m)	0.10	0.20
Density, (g/cm <sup>3</sup> )	0.92	0.90
Melting point, (°C)	160	180

50.00 mm. The test was carried out at controlled atmosphere of 22±2°C, at relative humidity of 60%. Testing was performed at 20 mm/mins of cross-head speed by using a 20 kN Universal Testing Machine (Shimadzu AGS Z, from Japan). For each loadings, about five samples were tested for data averaging purpose. For the case of this study, the tensile strength (TS), tensile modulus (TM) and the elongation at break (EAB) responses were analysed and presented, to represent the mechanical effects of various formulation of r-HDPE/r-PP blends.

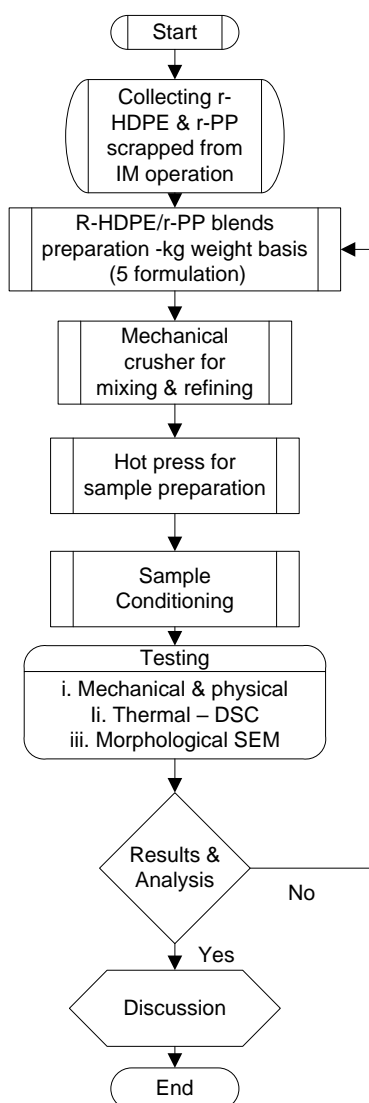
For hardness testing, the shore-D hardness test was performed for measuring the local indentation at soft hardness surfaces of r-HDPE/r-PP blend samples. About five (5) different points were taken from each sample for averaged data determination for three times of testing repetition. The shore-D hardness test measuring device was consisted of measurement unit with sensor and 360° anti-glare dial with an accuracy of 0.50 of hardness unit.

### 2.3 Differential Scanning Calorimeter (DSC)

The differential scanning calorimeter test was performed to understand the heat flow behaviour of tested r-HDPE/r-PP blend samples. The testing was performed by using the DSC machine set-up model Jade from Perkin Elmer. About 5.00 – 15.00 milligrams of blend samples was cut and stored at an inert aluminium metal pan for thermal analysis. The closed-trimmed aluminium pan was then placed inside the furnace and the temperature was raised up from controlled room temperature at 25°C into 200°C, at a heating rate of 10°C/mins. Performing this thermal analysis would give indication on miscibility state between the r-HDPE and r-PP phases within the r-HDPE/r-PP blends, by inspecting the variation of melting temperature (T<sub>m</sub>) peak produced at the temperature scan.

### 2.4 Fracture Surface Morphological Observation of r-HDPE/r-PP Blends

The fracture surfaces of the selected samples of r-HDPE/r-PP blend samples are based on the tensile strength experimental result that was observed under the Scanning Electron Microscope (SEM) observation. At first, the fragment of fracture surface was cut and mounted onto the stub with carbon tape before being coated with gold-palladium (Au-Pd) thin



**Fig 1.** Overall flow of the study

### 2.2 Mechanical and Physical Testing

The tensile test for r-HDPE/r-PP were performed in accordance to ASTM D638 – Type 1. Dimension of tensile test sample was monitored within ± 2.00 mm of thickness, overall width of 19.00 mm and gauge length at

conductive coating, using a sputter coater model Polaron E-1500, to eliminate the charging effects during the observation. The fracture surface images were captured by using Zeiss Evo VPSEM at 7.00kV accelerating voltage for 100x of magnification power at secondary electron detection. Distribution of r-HDPE and r-PP phase within the r-HDPE/r-PP blend sample was observed. From the fracture morphological observation, the structure-properties relationship for r-HDPE/r-PP blend was established.

### 3. Results and Discussion

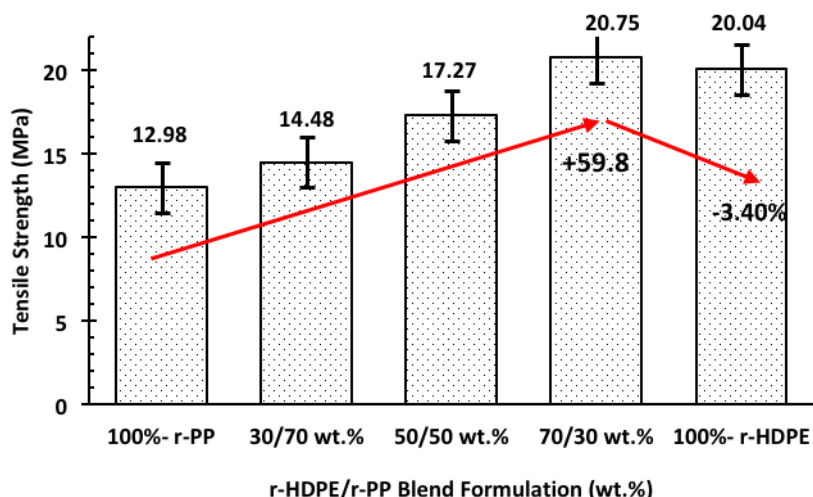
#### 3.1 Mechanical and Physical Properties of r-HDPE/r-PP Blends

The tensile strength (TS) performance of r-HDPE/r-PP blends was reported as in Figure 2. Increasing trend of TS was established with the increasing percentages of r-HDPE in the blend. However, this trend was sustained up to 70 wt.% of r-HDPE phase that are available within the r-HDPE/r-PP blend. The positive improvement in comparison with control sample of 100 wt% r-PP has reached about 59.8%. The sample with 100 wt.% of r-HDPE has experienced TS dropped about 3.40% than 70/30 wt.% r-HDPE/r-PP blend sample. The maximum TS performance as indicated by r-HDPE/r-PP blend sample with higher content of r-HDPE are caused by the stiffer characteristic of r-HDPE but not contributed by the good strength of PP component. The recycled sample of 100% r-PP shows massive strength reduction in comparison with virgin PP (refer Table 1). About 75.03% of this reduction is due to lack thermal stability properties of PP which experienced massive chain cutting or scission during their first cycle of IM operation. This situation has clearly explained the highest TS value of 70/30 wt.% r-HDPE/r-PP blend in term of their best tensile strength performance. This also tells that, presence of r-PP at smallest portion (30wt.%) are able to strengthen the blend but not at the highest portion. This situation was well explained by the TS reduction of 100% r-HDPE sample.

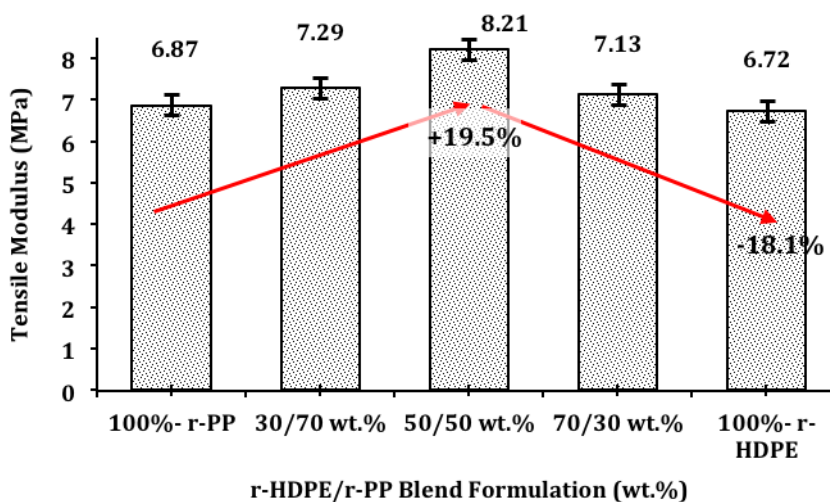
The following Figure 3 depicts the tensile modulus (TM) results of r-HDPE/r-PP blends at various combination ratio. Similar like the tensile strength plot, it was found that by increasing the r-HDPE content in the blend up to 50 wt.%, about 19.5 % of TM enhanced improvement has been established. However,

further increased the r-HDPE at 70 wt.% portion in the r-HDPE/r-PP blend had diminished the TM performance about 18.1% reduction in comparison with r-PP without r-HDPE sample. Decreasing pattern of TM at higher percentage of addition was justified by the lowest TM performance of r-HDPE at 100 wt.% at only 6.72 MPa. The performance of r-HDPE without r-PP was lesser than the performance of r-PP without r-HDPE. At balance proportion of r-HDPE and r-PP, the 50/50 wt.% r-HDPE/r-PP blend was at the peak performance of TM considering the advantage of r-PP that having higher thermal stability for second processing cycle and the advantage of r-HDPE that having good flexural modulus which reflect the stiffness attribute. Advantages from both r-HDPE and r-PP phases are well combined for this blend formulation ratio. The properties dropped at 70/30 wt.% r-HDPE/r-PP suggest the improper dispersion between r-HDPE and r-PP phases at higher content of r-HDPE, which limits their miscibility. Partial or limited miscibility able to promote early failure due to phase separation when the blend not able to sustain the load during the deformation or loading. Lack performance of r-HDPE at 100 wt% without the presence of r-PP was confirmed by the fracture morphology observation via SEM, which clearly found the presence of impurities or contamination that indicate un-melted r-HDPE at their matrix counterparts. These contaminations will act as a stress raiser or crack initiation point that lead into lowering the TM of produced sample.

Unusual performance of elongation at break (EAB) was recorded for the developed r-HDPE/r-PP blends at various formulation as available in Figure 4. The pattern of plots were exactly the same with TS performance. The highest EAB value was recorded by r-HDPE/r-PP blend sample at 70/30 formulation ratio, which similar with the higher TS performance. Partial miscibility between r-HDPE and r-PP at 70/30 ratio had responsible to induce good strength behaviour, but at the same time able to cause well interaction between macromolecules of both phases that allow for flexibility and ductility. Proper interaction at 70/30 improved EAB about +473% as compared than 100% r-PP. Good molecular interlocking between both r-PP and r-HDPE at this ratio had increase the interfacial adhesion between phases that would allow free chain mobility between interface.



**Fig 2.** Tensile strength of r-HDPE/r-PP blends.



**Fig 3.** Tensile modulus of r-HDPE/r-PP blends.

This finding, had supported the reduction result experienced by r-HDPE/r-PP blend at 70/30 due to inability to resist the deformation as portrayed by higher EAB at this ratio.

The variation of Shore-D hardness values of r-HDPE/r-PP blends are presented as in the following Figure 5. The hardness values was gathered by using Shore-D hardness tester. Although the fact that PP is stronger and more rigid than HDPE but the Shore-D findings for this recycled material blends was totally opposite from this fact since PP has higher hardness value than the other polymeric types [8]. Furthermore it was noticed that the Shore-D values for r-HDPE / r-PP blends had increased by the increase of r-HDPE in the formulation of about +2.30% at 70% of blend

ratio, but the hardness value was dropped into 0.16% without any existence of r-PP. From this plot it has been noticed that the highest value was recorded at 63.6 Shore-D hardness values for 70/30 r-HDPE/r-PP blend. The r-PP without the existence of r-HDPE would give the lowest value of Shore-D hardness. This observation further support the role of r-HDPE in enhancing some of the mechanical properties of r-HDPE/r-PP blends. This result was opposite from Salih et al. (2013) who stated that Shore-D values for HDPE/PP blend and LDPE/PP blend had experienced increase as the weight ratio of PP increased. This is due to the nature of PP which supposedly much stiffer than any other PE polymers [3].

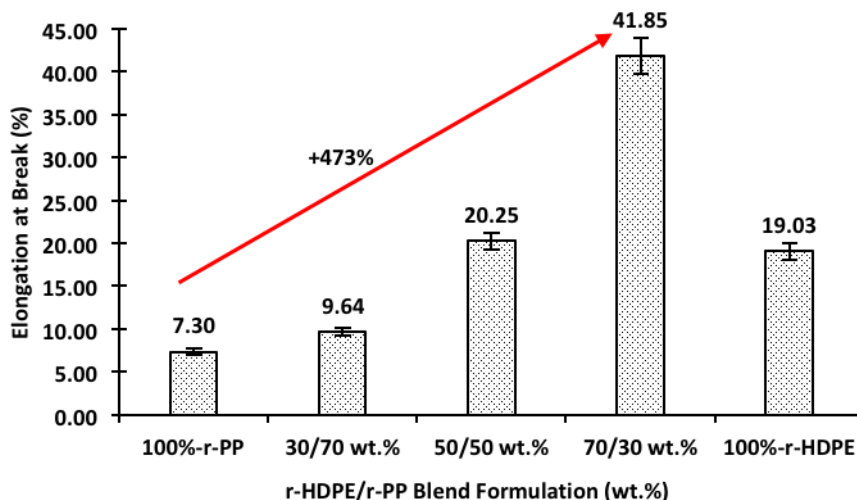


Fig 4. Elongation at break of r-HDPE/r-PP blends.

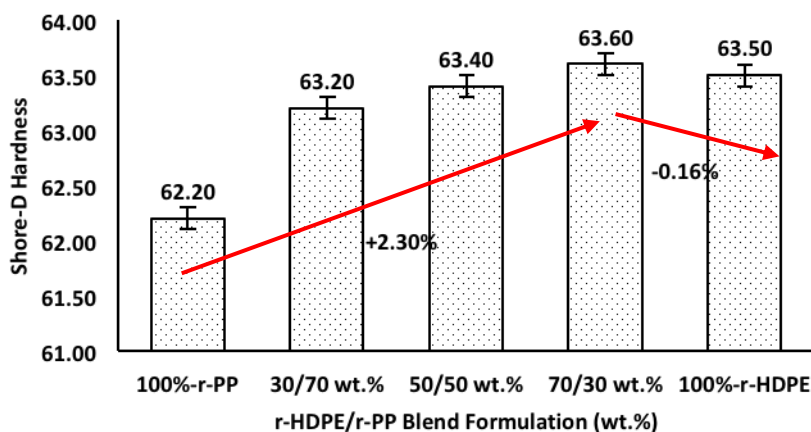


Fig 5. Shore-D hardness of r-HDPE/r-PP blends.

### 3.2 Heat Flow Analysis of r-HDPE/r-PP Blends by DSC

Heat flow analysis for thermal characterization of r-HDPE/r-PP blend was conducted by using the differential scanning calorimeter (DSC) machine. The thermogram for each sample was overlaid as in the following Figure 6. First of all, by looking at both control sample of 100% r-HDPE and r-PP, it was found that the thermogram having two major peaks at  $\sim 130^{\circ}\text{C}$  and  $\sim 160^{\circ}\text{C}$ . None of these peaks explained the melting temperature ( $T_m$ ) of both pure PP and HDPE component. The shifting into another  $T_m$  peak might due to recycling effect of virgin PP and HDPE from the IM process. Chain scission, contamination and macromolecules degradation due to recycling effect has contributed towards this

peak shifting phenomena. Comparison between blends consisting of 70/30, 50/50 and 30/70 r-HDPE/r-PP ratio has been made. Variation of blend ratio formulation has narrowing the  $\sim 130^{\circ}\text{C}$  melting peak. This observation has indicate the possibility of partial miscibility between the r-HDPE and r-PP blend phases. Partial miscibility at r-HDPE/r-PP blends has thermally explained the improvement experienced by some of the tested mechanical and physical attributes. It is well known that r-HDPE/r-PP is heterogeneous two-phase system, the components of which macromolecules has been separated into discrete phases [9, 10]. It seems that, there was limited or partial miscibility between the r-PP in r-HDPE, especially at lower contents of r-PP as shown in the following Figure 6. It can be seen that the

T<sub>m</sub> peak of r-HDPE in all blend compositions shifts to higher temperatures. In addition, the T<sub>m</sub> of r-PP shifts to lower temperatures due to the blending effects as also supported by Aumnate et al. (2016) [11]. This might be explained by less regular molecular structure and higher polydispersity index of r-HDPE

which resulted from oxidation and chain scission undergone during IM processing. It is reported by Madi (2013) in the case of incompatible blends, the melting point decreases since the noncrystallisable component retard the crystal growth, which leads to imperfect crystals [12].

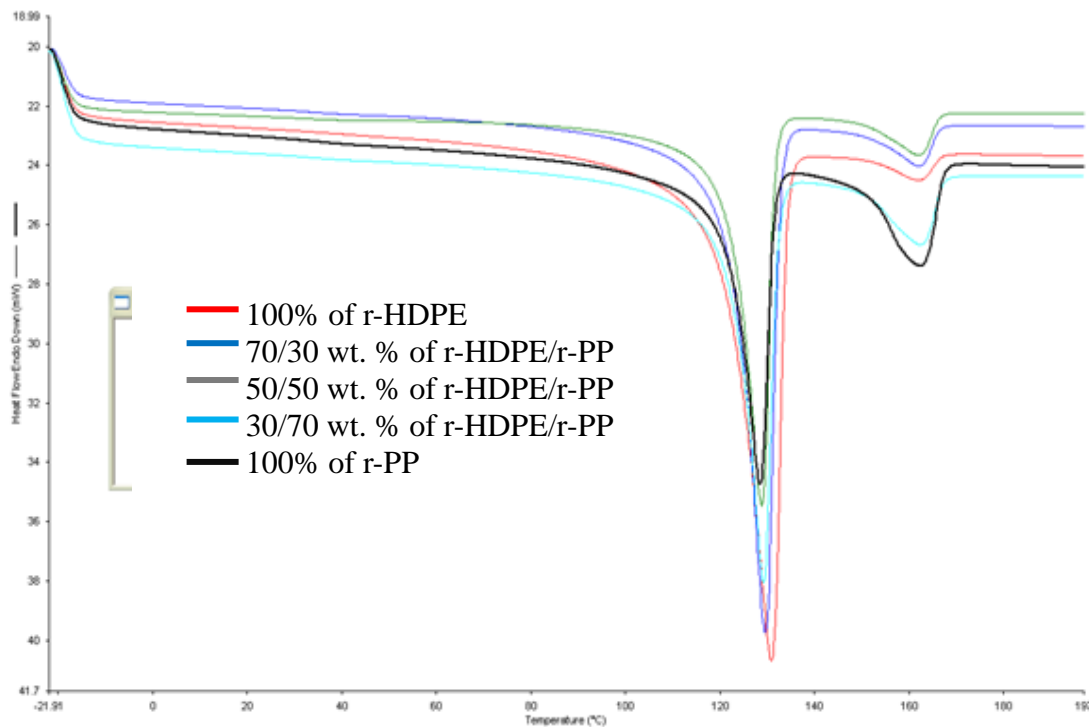
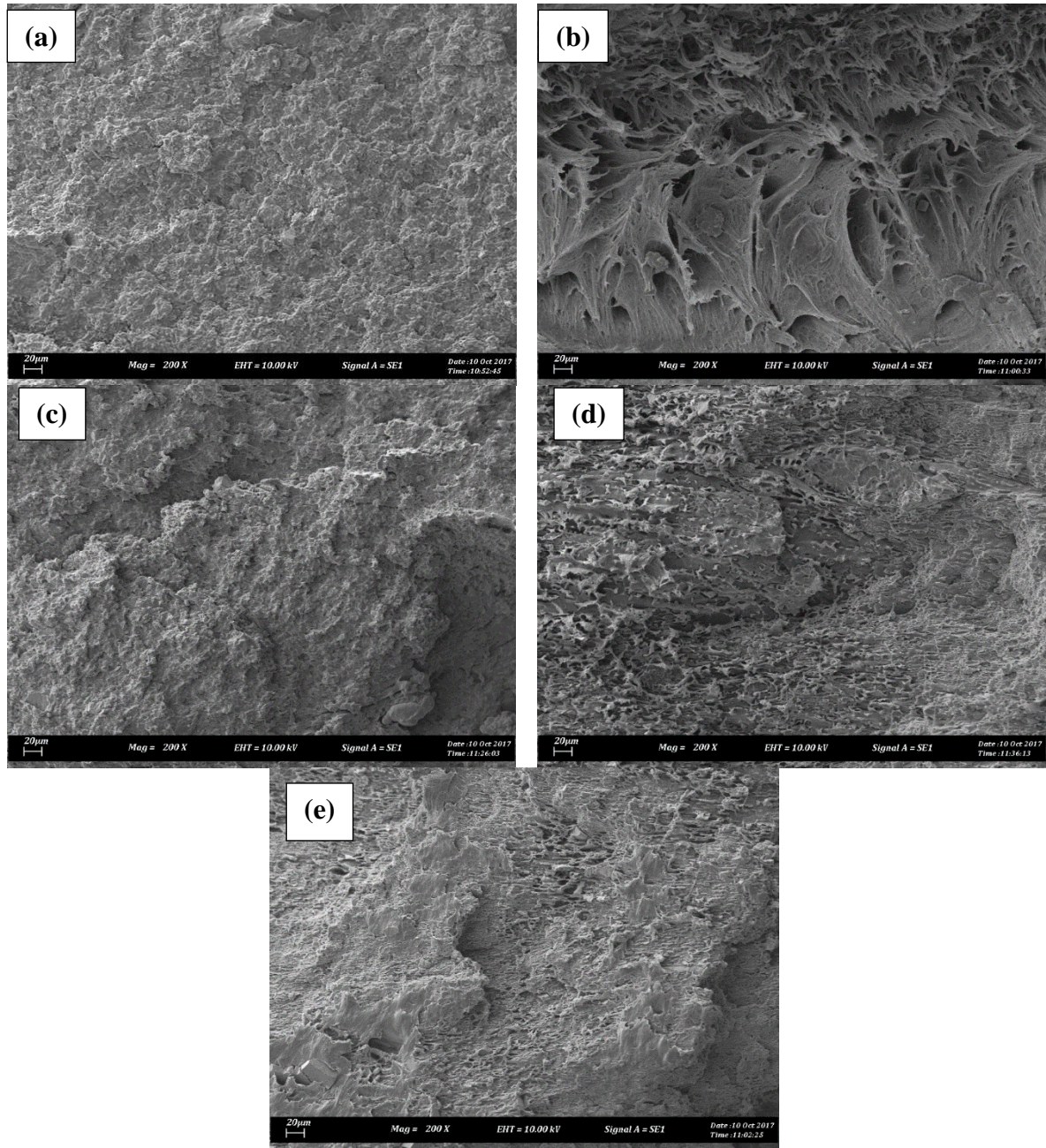


Fig 6. DSC thermogram of r-HDPE/r-PP blends.

### 3.3 Fracture Morphological Observation of r-HDPE/r-PP Blends

The following Figure 7 depicts the fracture morphologies of r-HDPE/r-PP blends consists of 100% r-PP, 100% r-HDPE, 70/30 wt.% of r-HDPE/r-PP blend, 50/50 wt.% of r-HDPE/r-PP blend and 30/70 wt.% of r-HDPE/r-PP blend samples at 200x magnification, respectively. For Figure 7(a) and 7(b) represent the r-PP and r-HDPE component without blend phase. It was clearly observed that both r-PP and r-HDPE possessed continuous morphology which tells compound homogeneity through hot pressing process. However, what make difference between these two micrograph are the roughness of the fractured surface. r-HDPE sample possessed rougher surface than r-PP which explaining some of the good mechanical attribute for r-HDPE as compared than r-PP, like the tensile strength, shore hardness and

EAB. r-HDPE exhibits ductile failure tearing mode while r-PP exhibits brittle fracture mode. This has explained the positive improvement on UTS, T<sub>m</sub> and EAB, when r-HDPE at the higher content in r-HDPE/r-PP blend. The SEM micrograph of rHDPE/rPP blend at 30/70 blend ratio has indicated rougher surface as shown in the following Figure 7(c). The 50/50 wt.% of r-HDPE/r-PP micrograph has clearly indicates that the r-HDPE and r-PP are experience incompatible phase separation towards each other. For example, if r-HDPE induces a change transformation in phase continuity, changing from a dispersed r-PP and r-HDPE phase as shown in the following Figure 7(a) and 7(b) to a more continuous phase as seen in the following Figure 7(c) and 7(d) are portrayed by an increase in mechanical properties of the respective r-HDPE/r-PP blend are observed.



**Fig 7.** Electron micrographs of r-HDPE/r-PP blends; (a) 100% r-PP; (b) 100% r-HDPE; (c) 70/30 wt.% of r-HDPE/r-PP blend; (d) 50/50 wt.% of r-HDPE/r-PP blend; and (e) 30/70 wt.% of r-HDPE/r-PP blend (at 200x magnification).

#### 4. Conclusion

In conclusion, combining r-HDPE/r-PP into hybrid blend are capable to improve the properties of primary r-HDPE and r-PP phase. Some of the mechanical and physical properties of r-HDPE/r-PP blend are better than r-PP and r-HDPE phase, such as the UTS, TM, EAB and Shore-D hardness. Partial miscibility condition that has been established at optimum portion of r-HDPE/r-PP formulation ratio greatly help in improving those attributes. r-HDPE phase play

a major role in sustaining the load, local indentation and even thermal stability for r-HDPE/r-PP blends. This was verified by the entire experimental findings gathered from this work. At the end, through this study, it can be said that the combination of recycled polymer offers good potential in replacing the use of virgin Delrin in this case for teaching and learning at IKTBN technical institution, in the future. This absolutely will give advantage into environmental sustainability, reducing raw



materials cost for teaching and learning, last but not least optimizing the in-house resources.

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