

THERMAL AND MECHANICAL BEHAVIOUR OF RECYCLED POLYPROPYLENE/POLYETHYLENE BLENDS OF REJECTED-UNUSED DISPOSABLE DIAPERS

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Article History: Received 15 August 2019; Revised 21 November 2019;
Accepted 15 December 2019

ABSTRACT: This paper presents the characterization of recycled plastic that derived from the rejected-unused disposable diapers containing polypropylene (PP) and polyethylene (PE), noted as r-PP/PE. The blends were tested for thermal, mechanical and morphological properties. Tensile test showed that the r-PP/PE is lower in strength and strain but higher for modulus in comparison to the v-PP/PE by 56%, 55%, and 2% respectively. For the flexural properties, the r-PP/PE also has lower in strength, strain, and modulus as 67%, 13%, and 77% respectively. Lower absorbed energy and impact strength was observed in r-PP/PE, 36% and 24% respectively compared to v-PP/PE. Thermal analysis revealed that the degree of crystallinity of recycled PP and PE was 19% and 20% lower than the virgin possibly due to thermal degradation during the process. Morphological examination revealed the present of impurity, phase separations and inhomogeneity were found in the r-PP/PE as compared to v-PP/PE that might contribute to their lower strength.

KEYWORDS: *Recycled-Unused Disposable Diapers; Recycled-Polymer Blend; Mechanical Properties; Thermal Properties; Morphological Properties*

1.0 INTRODUCTION

The sustainable development has become the main drive in utilizing renewable resources since it gave the significant impact of nature on human life, especially on its economic and social affairs. Some examples of renewable resource applications are in producing environmental friendly bio-composites from natural fibre as structural, automotive components and others [1-2]. Furthermore, sustainable development efforts are also heavily carried out to optimize existing resources through 3R (reduce, reuse, and recycle) initiatives. Recycling and reusing the scraps from industries could reduce the abundance of waste in landfills, reduce oil and energy consumption [3]. The use of recycled material as a resource is said as among the goals for sustainable consumption and production, and important for the realization of the close-loop economic option [4].

The polymer waste recycling can be done through mechanical separation, floating method, and chemical recycling methods [5-7], which could provide raw materials for product developments in related industries. Polypropylene (PP) and polyethylene (PE) are the most abundant raw materials used in plastic production. Their non-degradable nature has posed a disposal issue to the environment [9]. In this study, the authors tried to highlight one of the potential resources of polymer waste which is the polymer derived from rejected – unused disposable diapers (RUDD) containing PP and PE. Based on discussion with local Malaysian disposable diaper manufacturers, there are 600 pieces of diapers produced per minute, with 2% (12 pieces) of them being rejected due to quality defects. As reported by Colón et al., an unused disposable diaper with an average weight of 41 g was made up of 23% of plastic, consists of various thermoplastic polymers such as polypropylene (PP), polystyrene (PS) and elastic [8]. The calculation that can be made, there is 113 g/minute of unused diapers were rejected in the production line yielding approximately 4882 tons per month. Thus, the abundance of this potential resource can be recovered and perhaps being useful for product development.

Although the recycled polymers derived from the RUDD has the great potential, the report on their mechanical and thermal properties are still scarce. Thus, the aim of this study is to characterize the mechanical and thermal properties of the RUDD blends. Elemental analysis was carried out to identify the impurities that may contribute to alteration of their properties in comparison to the virgin blends. The findings from this study may reveal the potential recycled-polymer that derived from the RUDD as raw material for composite man.

2.0 MATERIALS AND METHODS

2.1 Materials

The received RUDD containing PP and PE is supplied by ZFH Industries Sdn. Bhd. (Klang, Malaysia) containing PP and PE contents with an average ratio of 70:30 wt.% (Figure 1). The commercial virgin PP (with density 1.48 g/cm³) and LLDPE (with gravity 0.9 g/cm³) were supplied by Lotte Chemical Titan and by Polyethylene Malaysia Sdn. Bhd., respectively.



Figure 1: Shredded RUDD as received

2.2 Sample Preparation

The recycle and virgin blends were compounded by using an internal mixer machine (ThermoHaake™, UK) at a temperature of 180°C, for 10 minutes at a rotor speed of 50 rpm. The blends were then compressed-moulded using a hot press machine (GoTech, Taiwan) in 200mm (width) x 200mm (length) x 3mm (thick) window frame mould using the parameters that were optimized in our previous study [9].

2.3 Thermal Analysis

Thermal analysis was conducted by a Perkin Elmer DSC thermal analyser. The samples were examined at a heating and cooling rate of 20°C/min in a nitrogen atmosphere from room temperature to 200°C and the cooled down to room temperature. The heating scan was repeated twice. The degree of crystallinity was obtained by using

$$\text{Crystallinity} = \frac{\Delta H_f^{obs}}{\Delta H_f^o} \times 100\% \quad (1)$$

where ΔH_f^{obs} is the observed heat of fusion values and ΔH_f^o is the heat of fusion values for 100% crystalline PP (209 J/g) or PE (140.6 J/g).

2.4 Mechanical Testing

The tensile test was carried out using a Universal testing machine (INSTRON, UK) (ASTM D638) at 2mm/min of crosshead speed. Five specimens in the form of dumbbell shape with a length of 165mm, the overall width of 18mm, and the width of a narrow section of 13mm were used. Extensometer with 25mm of gauge length was used for elongation detection and it was removed after 1% of elongation.

For the flexural test, five regular rectangular with a dimension of 125mm of length and 13mm of width were tested in accordance with ASTM D790. The tests were conducted at 2 mm/min of crosshead speed, and data including flexural strength and modulus were reported.

Ten rectangular samples with a length of 64mm and width 12mm were subjected to impact test (ASTM D256) using 2.65kg conventional impact tester. The impact energy was recorded and their strength was calculated by using

$$\text{Impact Strength (Jm}^{-1}\text{)} = \frac{\text{Impact Energy (J)}}{\text{Thickness of Material (m)}} \quad (2)$$

2.5 Morphological Analysis

The fracture morphology of the composite samples was analysed using EVO MA scanning electron microscope (Carl Zeiss, Germany). The analyses conducted at accelerating voltage of 10kV. All the surface fractures were gold coated using a mini sputter coater prior to the test. The energy-dispersive X-ray (EDX) was used for elemental identification.

3.0 RESULTS AND DISCUSSION

3.1 Thermal Analysis

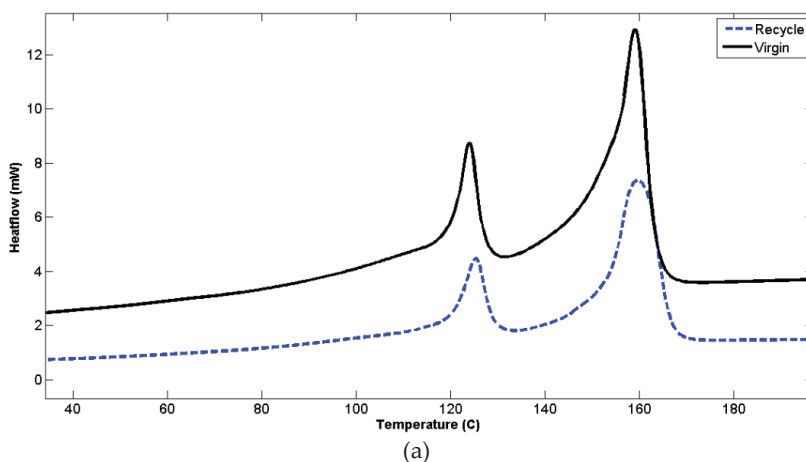
Table 1 and Figure 3 show the thermal properties of the PP/PE of the virgin and recycled blends. Two melting endotherms peaks corresponding to PP and PE phases, representing crystallization melting point (T_m) of the PP and PE in the blends, 162°C and 125°C respectively. Meanwhile, the T_m of PP and PE from the virgin blends are 160°C and 124°C. This similar melting during the first and second heating scans suggested that both blends has similar contents ratio of PP and PE. In addition, the presence of two melting peaks indicating the immiscible and incompatible of the blends [10-12]. The onset temperatures of melting and crystallization of individual PP and PE

are slightly lower in recycled compare to virgin blend suggesting that thermal degradation occurred and more severe in the recycled blends. This finding is similar as reported by Callister and Rethwisch [13].

The crystallization temperature (T_c) in the recycled blends is different as compared to the virgin blend. In both first and second cooling, there is only a single peak appeared for the recycled blends, meanwhile there are double peaks is detected in the virgin blend. The T_c for the recycled blends in both first and second cooling are the same, at about 109°C . The T_c of the virgin blend which are 115°C (PP phase) and 106°C (PE phase) for first cooling and the values for second cooling are 115°C and 107°C respectively, confirming the immiscibility condition that occurred in the virgin blend. Thus, it can be said that the recycle blend is more miscible during recrystallization compared to the virgin blend, and the recrystallization of PP took place in molten PE [14]. The degree of crystallinity of recycled PP and PE was 19% and 20% lower than virgin PP and PE, suggesting that thermal degradation has occurred in the recycled blends.

Table 1: Thermal characteristics of the PP/PE blends

| Blends | Element | Percent crystallinity (%) |
|---------------|---------|---------------------------|
| recycle blend | PP | 17.18 |
| | PE | 6.54 |
| virgin blend | PP | 21.53 |
| | PE | 10.81 |



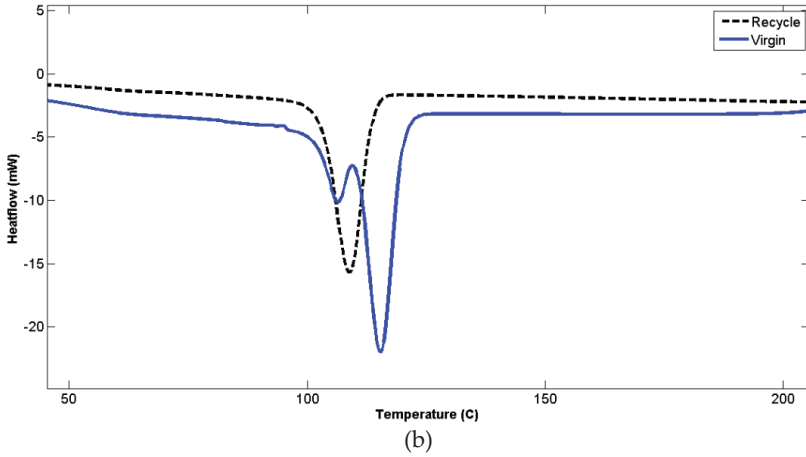


Figure 1: DSC heating curves and peaks showing the melting temperature of the recycled and virgin blends of PP/PE: (a) heating and (b) cooling

3.2 Tensile and Flexural Properties

The tensile properties of the blends were tabulated in Table 2. The average tensile strength and strain obtained for recycled-polymer blend were 9.4 MPa and 4.7% respectively, lower by 56% and 55% as compared to the virgin blends. Interestingly, the average modulus obtained by the recycled-polymer blend was 285MPa, 2% higher than virgin polymer blend, indicating both blends having similar rigidity. The SEM images in Figure 4 (a) indicated that the recycled-polymer blend has lower immiscibility, poor PP and PE dispersion, and insufficient interfacial adhesion compared to virgin blend. The poor homogeneity and the presence of the impurities may contribute to their mechanical degradation. Meanwhile, Figure 4 (b) showed the virgin PP droplets, and polymer stretched when subjected to tensile load indicated the ductile nature of the polymers similar to those reported by Bertin et al. [15].

Table 2: Tensile properties of recycled and virgin PP/PE blends

| Mechanical properties | Tensile | | | Flexural | | |
|-----------------------|----------------|------------|---------------|----------------|------------|---------------|
| | Strength (MPa) | Strain (%) | Modulus (MPa) | Strength (MPa) | Strain (%) | Modulus (MPa) |
| recycled blend | 9.4 (0.6) | 4.7 (0.6) | 285.2 (9) | 12.1 (0.7) | 6.8 (0.8) | 231.1 (26) |
| virgin blend | 21.5 (2.9) | 10.4 (1.1) | 280.6 (47) | 36.6 (3.0) | 7.8 (3.2) | 998.4 (127) |

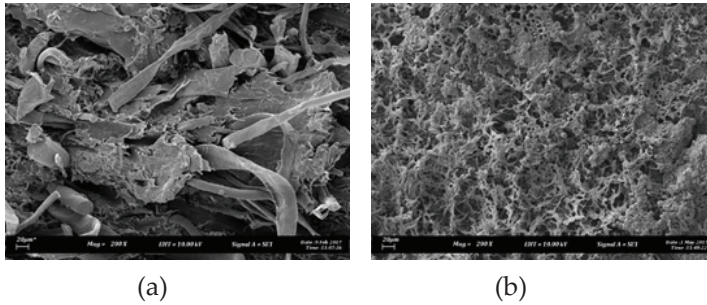
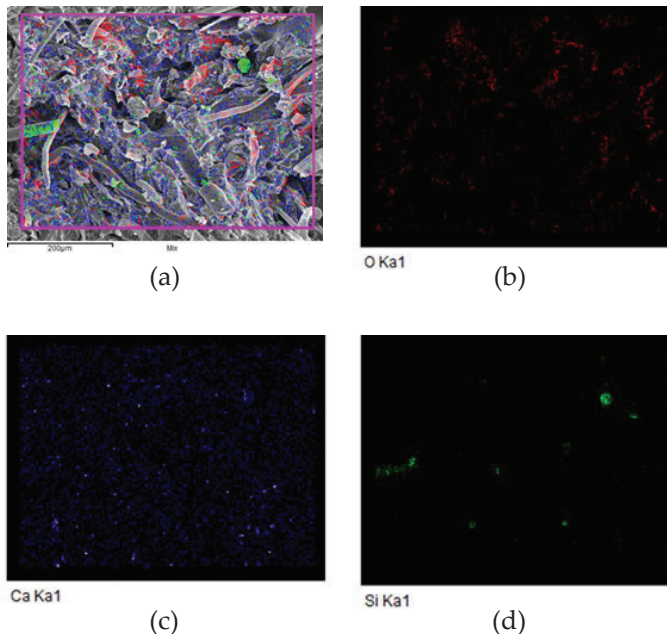


Figure 4: Fracture surface for PP/PE blends: (a) recycled and (b) virgin (scale bar = 20 μm)

The identified impurities that presence inside the recycled blends was in the form of thin-strip and rounded-shape. These impurities promoted discontinuity of the impurities during tension load is applied. Through EDX mapping, the comparison of the elements that exist in both blends can be done so that the specific impurities can be identified. Figure 5 showed the EDX mapping for recycled polymer blend which revealed that the blend has elements of oxygen, calcium, silica, titanium, and carbon, whereas the EDX mapping for virgin polymer blend showed only elements of carbon and silica, showed in Figure 6. The titanium and oxygen in recycled polymer blend may originate from titanium dioxide (TiO_2) which used as the white colouring and opacifier for the plastic products such as disposable diapers [15]. The other elements may be used as the absorbent agent material [16].



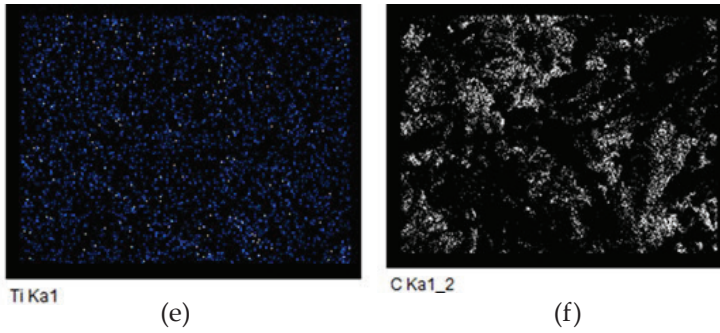


Figure 5: Fracture surface of (a) recycled PP/PE blend and the corresponding EDX elemental mappings: (b) oxygen, (c) calcium, (d) silicon, (e) titanium and (f) carbon (scale bar = 200 μm)

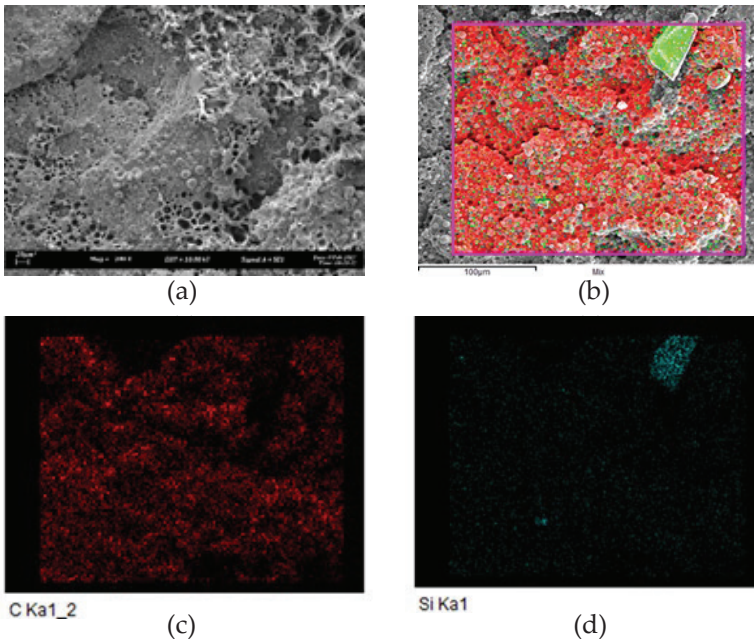


Figure 2: Fracture surface microstructure of (a)-(b) virgin PP/PE blend with the corresponding SEM-EDX elemental mappings of: (c) carbon and (d) silicon (scale bar = 20μm)

The flexural strength, strain and modulus of recycled polymer blend obtained were 12.1 MPa, 6.8%, and 231 MPa, which 67%, 13% and 77%, lower than the virgin blends. The lower strength of the recycled blends may due to loss of chain orientation caused by the recycling process [13]. The flexural behaviour of the blends might be affected by the degree of crystallinity similar to tensile behaviours. Higher exposure of heat during the recycling process may damage the long

chains of the polymers and reduced the crystallinity as shown by the thermal analysis. Virgin polymer, contains larger crystalline regions and more molecular chains. Higher crystallinity also may increase the extent of intermolecular secondary bonding between adjacent chain segments resulting in more ductile nature. During flexural loading, the blend with a lower degree of crystallinity display larger deterioration compared to a higher degree of crystallinity [17].

3.3 Impact Properties

The impact energy and impact strength of the virgin blend is higher compared to the recycled blend as expected, in which the virgin blend required 1.18 J impact energy to break with the strength to withstand impact 409 J/m (Table 3). Meanwhile, the recycled blends required a small amount of energy to break as 0.90 J and impact strength of 309 J/m, indicating that the materials become more brittle. This finding is in correlation with their tensile and flexural behaviours.

Table 3: Impact properties of the recycled and virgin PP/PE blends

| Mechanical properties | Impact | |
|-----------------------|-------------|----------------|
| | Energy (J) | Strength (J/m) |
| recycled blend | 0.90 (0.25) | 309 (46) |
| virgin blend | 1.40 (0.14) | 409 (228) |

During impact, the materials could fail either under brittle fracture, ductile fracture, yielding, and slight cracking failure [3, 17]. From the observation, both recycled and virgin blends displayed failure mode of yielding, where the specimen portrayed obvious and permanent stress whitening but no apparent cracking was observed.

4.0 CONCLUSION

The comparison towards virgin polymer blend on mechanical properties showed the recycled polymer blend having a weaker performance for tensile, flexural, and impact, yet only higher in tensile modulus. The poor mechanical performance of recycled polymer blends is due to a lower degree of crystallinity and immiscibility between PP and PE phases compare to virgin blends. Nevertheless, their mechanical properties are still within competitive value and they could enhance by blending with virgin polymers or as matrix in composite materials but at a fraction cost of the virgin material.

ACKNOWLEDGMENTS

The authors acknowledge the financial and support from the Universiti Teknikal Malaysia Melaka (PJP/2015/FKM(3A)/S01409), ZFH Industries Sdn. Bhd. For the raw materials and Centre of Advanced Research on Energy (CARE) and Advanced Manufacturing Centre (AMC) for using their laboratory and equipment.

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