Mechanical properties of wood polymer composites made of sawdust waste and recycled high density polyethylene

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Abstract
This work investigated the mechanical properties of wood polymer composites (WPCs), which were made using recycled high density polyethylene (HDPE) and sawdust waste. Degraded HDPE was used as a coupling agent. Different blends of virgin (vPE) and recycled (rPE) poly ethylene's with various ratio (100/0, 75/25, 50/50, 25/75 and 0/100) were prepared in order to be used as matrices. PE blends and composites were prepared by melt-mixing technique using mini twin-extruder. Mechanical properties such as ultimate tensile strength (UTS), impact strength and Shore hardness were determined for vPE, rPE, PE blends and PE composites. The mechanical properties of PE blends and composites were mainly depended on the composition of its matrices. Presences of degraded HDPE as a coupling agent played an important role on the mechanical properties of WPCs. Increasing the rPE content in the blend composition displayed decrease in the UTS and impact strength properties. The incorporation of only sawdust into vPE, rPE and PE blends matrices caused reduction in UTS and impact strength properties and an increase in Shore hardness. UTS, impact strength and Shore hardness for all WPCs were improved when 5% degraded PE was added. UTS and Shore hardness for composites with 5% degraded PE were higher than that of its matrices. The properties of PE composites were better than that of PE blends, which suggesting that using rPE to attain blends with vPE and then

making composites appears to be more preferable than making only PE blends.

Key words: recycled polyethylene, coupling agent, mechanical properties, sawdust.

الملخص

تم في هذه الدراسة التحقق من الخصائص الميكانيكية لبوليمرات مركبة تم تصنيعها باستخدام البولي إيثيلين عالي الكثافة (HDPE) المعاد تدويره ونشارة الخشب. كما تم استخدام HDPE التألف (Degraded HDPE) كمادة رابطة (Coupling agent). في البداية تم تحضير مخاليط من البولي إيثيلين (Polyethylene blends) البكر (vPE) والمعاد تدويره (rPE) بنسب مختلفة (0/100 ، 25/75 ، 50/50 ، 75/25 و 100/0) وذلك لاستعمالها كمادة أساس لتحضير بوليمرات مركبة. بعد ذلك حضرت البوليمرات المركبة باستخدام vPE و rPE والمخاليط البوليميرية ونشارة الخشب باستخدام آلة بثق مزدوجة. إجريت بعد ذلك دراسة الخواص الميكانيكية مثل مقاومة الشد القصوى (UTS) ومقاومة الصدمة والصلادة لكل من vPE وrPE والمخاليط البوليميرية والبوليمرات المركبة. اعتمدت الخواص الميكانيكية للمخاليط البوليميرية والبوليمرات المركبة بشكل أساسي على نسب المواد الداخلة في تركيبها. كما لعب HDPE التألف دورًا مهمًا في تحسين الخواص الميكانيكية للبوليمرات المركبة عند استعماله كمادة رابطة. أدت زيادة محتوى rPE في تركيبة المخاليط البوليميرية إلى انخفاض في UTS ومقاومة الصدمة. أدى أضافة نشارة الخشب وحدها لكل من vPE و rPE و المخاليط البوليميرية إلى انخفاض UTS ومقاومة الصدمة وزيادة الصلادة. هذه الخصائص (UTS ومقاومة الصدمة والصلادة) لجميع البوليمرات المركبة تحسنت عند إضافة 5% من HDPE التألف كمادة رابطة. UTS ومقاومة الصدمة والصلادة للبوليمرات المركبة المحتوية على 5% من HDPE التألف كمادة رابطة وجدت بأنها أعلى من مادة الاساس أو نظيرتها في المخاليط البوليميرية. بشكل عام خصائص البوليمرات المركبة كانت أفضل من المخاليط البوليميرية، مما يشير إلى أن استخدام rPE لتحضير بوليمرات مركبة مع vPE يكون

أفضل من استخدامها لصنع مخاليط البوليميرية في حال استعمالها في تطبيقات تعتمد على الخصائص التي تم دراستها في هذه الدراسة.

Introduction

The use of solid waste and recycled materials is becoming increasingly and vitally important. The utilization of recycled materials has become a key factor in making the world sustainable in the future. Recycling of waste materials saves natural resources, saves energy, reduces solid waste, reduces air and water pollutants and reduces greenhouse gases. There are a wide range of materials that can be recycled. These consist of obvious materials, including plastic, paper, glass, wood and metal. Plastics are the most commonly recycled materials in many countries. This is because the vast majorities of plastics waste are disposed of in non-environmentally friendly ways, resulting in polluted oceans, overextended landfills and ecological damage. Moreover, plastics waste can take hundreds of years to biodegrade. Recycling is the reprocessing of plastic waste into new and useful products.

There is lot of scope to utilize the plastic waste in combination with other materials towards improvement of recycling of plastics [1]. For example, the use of recycled and waste plastics has been considered for producing wood plastic composites (WPCs). WPCs are very promising and sustainable green material to achieve durability without using toxic chemicals. It was originally discovered in Italy in 1970s, and popularized in North America in 1990s [2]. The term WPCs refers to any composites that contain plant fiber and plastics. In fact, WPCs support the use of both recycled plastics and wood [3]. When compared with individual materials, WPCs may also offer more better properties, consistent performance, lower production costs, and create an avenue for the utilization of renewable resources [4]. In this trend, intensive researches have focused on the utilization of waste materials in the development of WPCs [5-9]. The majority of researches conducted on the field of WPCs clearly revealed that the recycled plastic and wood wastes can be used as a potential raw material to make composite products that can replace plastics in many applications, which contributing to circular economy and provide the above-mentioned benefits of recycling of waste materials. It is important to declare that recycled plastics of good quality would cost 60-70% of the price of virgin plastics but this value drops as properties are compromised from repeated recycling [10]. Only 14-18% of plastic waste is formally recycled, as a global average, and the percentage is much lower in some countries [11]. This indicates that the recycling rates of plastics in worldwide are too low. However, recycling only part of the solution to the plastic pollution crisis not a complete solution.

In this work, the mechanical properties of WPCs made with completely waste materials were investigated. These composites were made from sawdust waste and recycled polyethylene (PE) as well as degraded PE as coupling agent. From the knowledge and information related to polymer composites in most of the literature, the fabrication of WPCs from sawdust waste and recycled PE would technically feasible; however, the use of degraded PE like coupling agents could further enhance the properties of WPCs. For example, degraded PE was used successfully as a coupling agent in the production of WPCs [12-13].

Experimental work

Material

Recycled HDPE (rHDPE) were collected from local recycling storehouse located in Zawia, Libya. rHDPE were washed multiple times before chopped and used. Virgin HDPE (vHDPE) were purchased for Assahra Co & Ltd (Turkey). The melt flow rate (MFI) and density values of vHDPE were ~ 1 g/10 min (ISO 1133, 190 °C, 2.16 Kg) and 0.941-0.959 g/cm^3 (ISO 1183), respectively). rHDPE and vHDPE were used as a matrix separately and as a blend. Degraded HDPE was used as a coupling agent. Degraded HDPE was obtained by exposing the rHDPE in air oven for a period of 60 days at 100 °C.

Sawdust was collected from a local carpentry shop located in Zawia, Libya and used as a reinforcing filler. Sawdust can be considered as a mixture of different wood types. Sawdust was sieved to remove the impurities and passed through different sieves size to get particle size of ~ 212 μm.

Blend preparation

rHDPE/vHDPE blends with varying ratio (25/75 (named B1 as shown in Table 1), 50/50 (named B2 as shown in Table 1), and 75/25 (named B3 as shown in Table 1)), were prepared by melt-mixing technique using mini twin-extruder (Haake mini CTW twin screw) with average screw speed of 90 rpm., and the barrel temperatures set on 200 °C.

Composite preparation

Before composite preparation, sawdust was oven dried for 24 h at 90 °C. Composites were prepared by mixing individually rHDPE, vHDPE and rHDPE/vHDPE blends with sawdust (30%) and degraded HDPE (0 and 5%) using rotor ultra-centrifugal mill ZM 200. The final mixing was carried out using Haake mini CTW, twin screw with average screw speed of 90 rpm. and the barrel temperatures of 200 °C. The extruded materials were cooled in air and then granules to small pieces. All specimens for Tensile strength, impact strength and Shore hardness were prepared using injection molding (Explore 12ml). The prepared composites and their abbreviations are shown in Table 1.

Table 1. Abbreviations and compositions of the composite used in this study.

Nº	Composite code	Blend composition, %	Sawdust, %	Coupling agent, %
1	**vPE**	**100**	**0**	**0**
2	**vPE30**	**70**	**30**	**0**
3	**vPE30/5**	**65**	**30**	**5**
4	**B1**	**100**	**0**	**0**
5	**B1/30**	**70**	**30**	**0**
6	**B1/30/5**	**65**	**30**	**5**
7	**B2**	**100**	**0**	**0**
8	**B2/30**	**70**	**30**	**0**
9	**B2/30/5**	**65**	**30**	**5**
10	**B3**	**100**	**0**	**0**

11	**B3/30**	**70**	**30**	**0**
12	**B3/30/5**	**65**	**30**	**5**
13	**rPE**	**100**	**0**	**0**
14	**rPE30**	**70**	**30**	**0**
15	**rPE30/5**	**65**	**30**	**5**

Characterization

Determination of mechanical properties

The ultimate tensile strength (UTS) was determined using Shimadzu universal testing machine. Tensile test was performed at room temperature. Four specimens (73mm x 4mm x 2mm) were tested for each sample. The charpy impact test was carried out to determine the impact strength of the rHDPE, vHDPE and all composite materials using (CEAST Resil Impactor tester), with impact energy of 15 J. The specimens for impact test were prepared and notched according to ISO 179. Five specimens were tested for rHDPE, vHDPE and each composite. The hardness of molded HDPE, LDPE and their blends were determined using a Shore D durometer (RayRan) in accordance with ISO 868:2003. Hardness value for each sample is an average of 10 measurements.

Results and discussion

Mechanical properties results

UTS, impact strength and shore hardness results are shown in Table 2. It is important to note that the standard deviations are given in parentheses next to the values of the mechanical properties. In general, the UTS and impact strength of vPE were greater than the that of rPE and PE blends. As expected, increasing the content of rPE in the blend composition displayed decrease in the UTS and impact strength properties of vPE. As can be seen in Table 2, blending vPE with rPE did not produce significant changes in Shore hardness. These results, however, indicate that the incorporation of rPE into the vPE matrix did not cause any improvement in the UTS, impact strength and shore hardness properties. In general, recycled polymers tend to have weaker mechanical properties than their virgin counterparts, since they may suffer degradation from heat, mechanical stress, oxidation or ultraviolet radiation during their lifetime and reprocessing [14]. Even polymers with short lifetimes are susceptible to degradation [15].

Table 2. Mechanical properties of vPE, rPE, PE blends and PE composites.

Nº	Composite code	Ultimate tensile strength, MPa	Impact strength, KJ.m^{-2}	Shore hardness
1	**vPE**	**56.05 (1.4)**	**52.6 (5.3)**	**60.05 (6.6)**
2	**vPE30**	**39.04 (1.5)**	**14.5 (0.6)**	**62.20 (8.9)**
3	**vPE30/5**	**65.74 (2.1)**	**16.9 (3.5)**	**63.75 (7.5)**
4	**B1**	**35.17 (1.1)**	**21.4 (5.5)**	**61.40 (4.6)**
5	**B1/30**	**32.10 (0.1)**	**13.1 (0.4)**	**61.40 (12.6)**
6	**B1/30/5**	**50.44 (1.6)**	**15.0 (0.6)**	**61.90 (13.5)**
7	**B2**	**28.88 (2.8)**	**20.8 (1.7)**	**60.60 (4.9)**
8	**B2/30**	**27.78 (0.9)**	**18.4 (1.1)**	**62.95 (14.8)**
9	**B2/30/5**	**29.16 (0.8)**	**20.3 (1.6)**	**63.15 (7.8)**
10	**B3**	**26.92 (0.6)**	**20.2 (0.6)**	**60.60 (11.9)**
11	**B3/30**	**25.90 (0.5)**	**19.1 (0.2)**	**62.40 (11.7)**
12	**B3/30/5**	**28.15 (0.6)**	**19.6 (1.1)**	**64.70 (9.2)**
13	**rPE**	**35.27 (2.2)**	**42.2 (1.0)**	**61.95 (7.6)**
14	**rPE30**	**34.84 (1.4)**	**23.0 (0.6)**	**64.40 (12.4)**
15	**rPE30/5**	**37.82 (2.2)**	**24.3 (1.1)**	**65.75 (12.5)**

The incorporation of only sawdust to vPE, rPE and PE blends matrices caused reduction in UTS and impact strength properties. Indeed, this can be attributed to the weak interfacial adhesion caused by the low compatibility between hydrophobic PE and hydrophilic wood fiber. The low compatibility between the two components caused in poor stress transfer from the vPE or rPE or their blends to filler, results in composites with lower mechanical properties [14]. According to Ezenkwa et. al. [17] weak fiber-matrix compatibility causes weak interfacial adhesion resulting in decreased composite tensile strength, elongation at break and impact strength. On the other hand, incorporation of only sawdust to vPE, rPE and PE blends matrices resulted an increase in Shore hardness. The increase in hardness can be due to the reinforcement effect of the filler. This happened because when the wood sawdust incorporated into polymer matrix would conduct in a reduction of plasticity and flexibility of the polymer chains, consequently the composite become more rigid [18].

UTS, impact strength and Shore hardness for all the composites were improved when 5% degraded PE was added as a coupling agent (in comparison to composites without coupling agent). This means that using degraded PE as a coupling agent could improve the adhesion between hydrophobic polymer matrices (vPE, rPE and all the blends) and hydrophilic wood fiber (sawdust), which confirm previously published results [12-13]. As it be seen in Table 2 and Figures 1 and 2, UTS and Shore hardness for composites with 5% degraded PE were higher than that of its original matrices. For example, vPE30/5 (composite with 30% saw sawdust and 5% coupling agent) exhibited higher UTS and Shore hardness than that of vPE. The same trend was observed by rPE composites and composites made with PE blends.

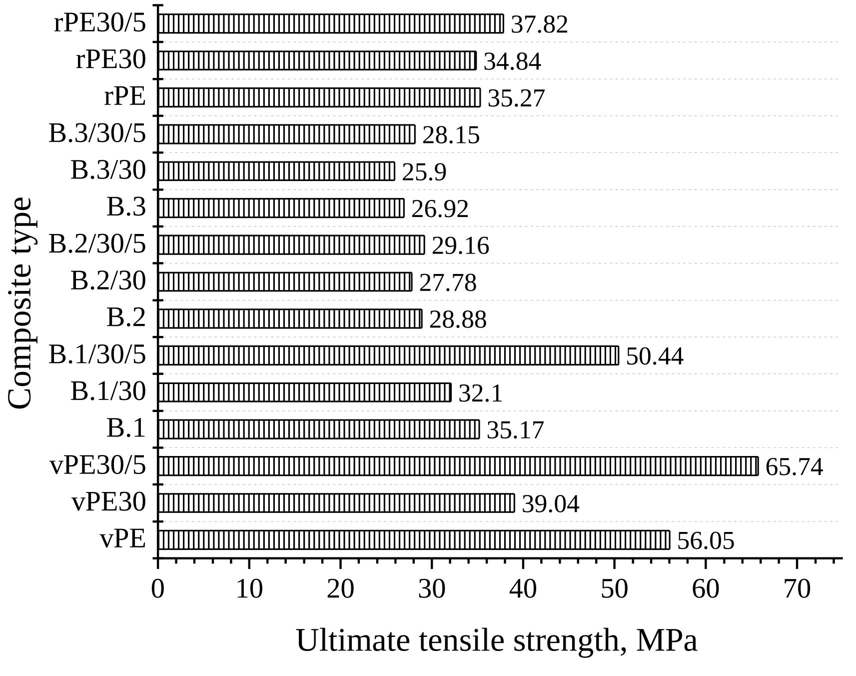

Figure 1. Ultimate tensile strength of vPE, rPE, PE blends and PE composites.

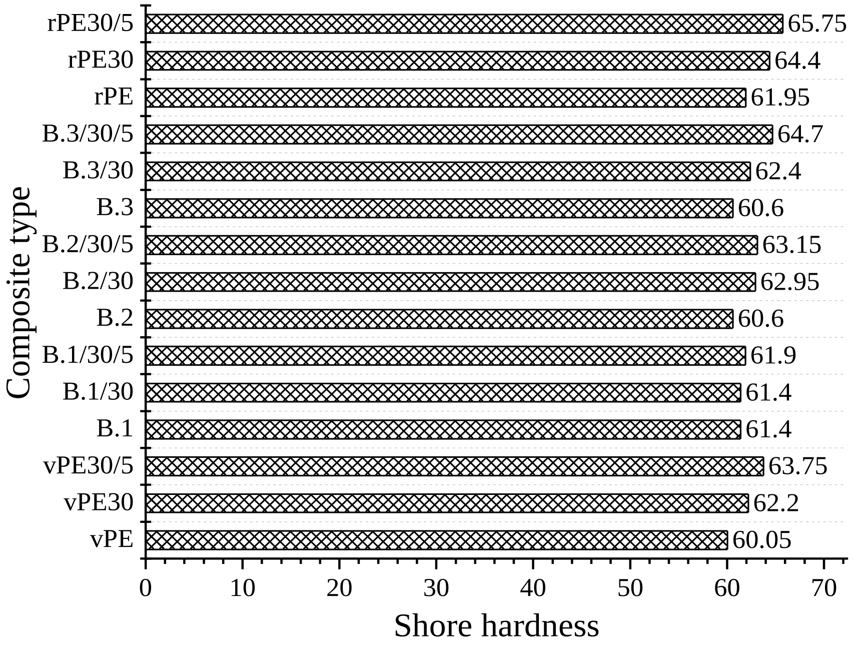

Figure 2. Shore hardness of vPE, rPE, PE blends and PE composites.

On the other hand, although the impact properties of all composites were improved by the addition of 5% degraded PE as a coupling agent, the impact strength values of these composites still lower than that of its original matrices. In other words, the impact strength values of vPE30/5, B1/30/5, B2/30/5, B3/30/5, rPE30/5 were higher than that of vPE30, B1/30, B2/30, B3/30, rPE30, respectively. The impact strength values of later mentioned composites were lower than that of vPE, rPE and their blends, as shown in Figure 3.

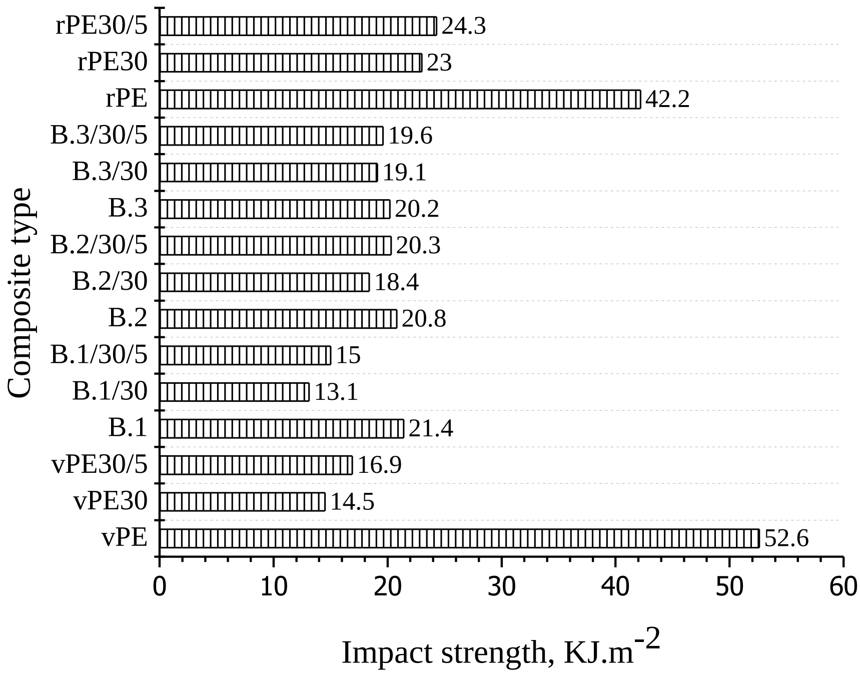

Figure 3. Impact strength of vPE, rPE, PE blends and PE composites.

It is important to know that recycled polymers are matched with potential products based on their strengths and weaknesses [14]. The utilization of rPE for producing WPCs, which conducted in this study has provided critical insight in finding their recyclability potential and their suitability to be used again. Looking closer at the results in Figure 4, the properties of PE composites were better than that of PE blends. This may suggest that using rPE to attain blends with vPE and then make composites appears to be more preferable than making only PE blends. Generally, recycled plastics must have the appropriate processing characteristics and mechanical properties to be reprocessed into the desired application [19].

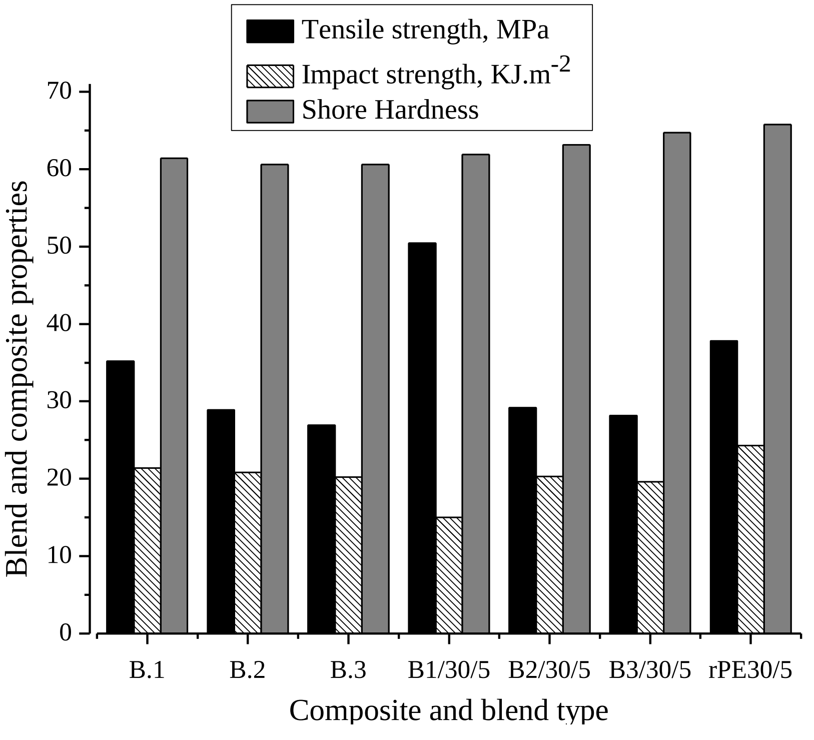

Figure 4. Comparison of the properties of PE blends and composites.

As it is well known, recycling plastic wastes may conserves energy, reduces pollution, and conserves natural resources. The same concepts may be applied on the utilization of sawdust, although it is green waste. The utilization of sawdust in many sectors of industry greatly improve the environment and economy. It should be considered as a material with a lot of promise. This is because sawdust could be put to useful agricultural, building and chemical uses. Sawdust is therefore no longer a waste material but an industrial material of use [20]. Moreover, the utilization of degraded plastics as a coupling agent in the flied of WPCs definitely provides more environmental and economical benefits to the production of these composites. However, these composites could be applicable for applications such as furniture, consumer goods sectors, in addition to other potential areas such as automotive, construction, and building.

Conclusion

This study investigated the mechanical properties of WPCs based on recycled and virgin HDPE using sawdust as a reinforcement and degraded PE as a coupling agent. The mechanical properties of composites were compared to those of vPE, rPE and vPE/rPE blends. The results indicated that the fabrication of WPCs from sawdust and HDPE waste was technically feasible; however, the use of degraded HDPE as a coupling agents could be further enhanced the properties of WPCs. The following conclusions can be drawn under the light of above results:

- UTS and impact strength of vPE were greater than the that of rPE and PE blends. Increasing the content of rPE in the blend composition displayed decrease in the UTS and impact strength properties of vPE.
- Blending vPE with rPE did not produce significant changes in Shore hardness compared to single vPE or rPE.
- The incorporation of only sawdust to vPE, rPE and PE blends matrices caused reduction in UTS and impact strength properties due to the weak interfacial adhesion caused by the low compatibility between hydrophobic PE and hydrophilic wood fiber. On the other hand, Shore hardness were increased by the incorporation of only sawdust.
- UTS, impact strength and Shore hardness for all the composites were improved when 5% degraded PE was added as a coupling agent.
- UTS and Shore hardness for composites with 5% degraded PE were higher than that of its original matrices.
- The properties of PE composites were better than that of PE blends, which suggesting that using rPE to attain blends with vPE and then making composites appears to be more preferable than making only PE blends.

- Studies in field of the utilization of recycled plastics must be directed to provide outcomes help to allow scientists and industrial to expect the properties and performance of the materials which produced from the available plastic wastes. This is because recycled plastics must have the appropriate processing characteristics and mechanical properties to be reprocessed into the desired application.

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