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Mechanical Properties of Recycled Polyethylene Terephthalate/ Polycarbonate/ Methylene Diphenyl Diisocyanate (r-PET/PC/MDI) Composite

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ABSTRACT

The present study aimed to investigate the mechanical properties of recycled Polyethylene Terephthalate (r-PET) and Polycarbonate (PC) composites, with the incorporation of Methylene Diphenyl Diisocyanate. The primary objective was to identify the optimal composition for achieving superior mechanical performance. The composition under examination comprises four ratios, namely r-PET/PC1 (20/80), r-PET/PC2 (40/60), r-PET/PC3 (60/40), and r-PET/PC4 (80/20), expressed as weight percentages. The concentration of MDI utilized in the experiment was maintained at a constant level of 0.9% based on weight. The r-PET/PC/MDI composite underwent blending using a Brabender Plastograph prior to being introduced into the injection moulding process for sample production. The mechanical properties can be ascertained through the analysis of data acquired through mechanical testing, encompassing Tensile tests, Flexural tests, and Impact tests. The findings indicate that there is an inverse relationship between the percentage of r-PET in the blend and the tensile strength, with an increase in r-PET resulting in a decrease in tensile strength. Conversely, there is a positive relationship between the percentage of r-PET and Young's modulus, with an increase in r-PET leading to an increase in Young's modulus. The flexural strength and flexural modulus exhibit variations as the r-PET content increases in the r-PET/PC/MDI blend. Lastly, it can be observed that the impact strength reduces as the proportion of r-PET in the blend increases. The findings of this experiment indicate that the composition of r-PET/PC1 yielded the most favourable results. The composition under investigation demonstrated the greatest recorded tensile strength, measuring 22.30 MPa, as well as the second highest flexural strength, measuring 119.77 MPa. However, the impact strength of r-PET/PC1 exhibited the greatest value compared to the other samples, measuring 0.32J. In summary, the findings indicate that the mechanical characteristics of the r-PET/PC/MDI mix can be improved through the optimization of PC content, hence enhancing the features of r-PET.

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1. Introduction

Land pollutants are defined as any form of waste that has the potential to inflict harm or have detrimental effects on the soil, groundwater, or overall land conditions. Waste management, particularly the disposal of rubbish, has emerged as a prominent topic of global discourse [1]. Rubbish typically consists of desiccated substances such as paper, glass, fabrics, and notably, plastic items. There exist multiple categories of plastic materials that serve as a significant contributor to the problem of land pollution. The aforementioned form of pollution has the potential to impact all facets of existence, including the imperilment of species [1,2]. As a result, it is imperative to implement effective management strategies for plastic trash in order to mitigate the extent of land pollution and its associated detrimental effects.

One of the most straightforward methods for addressing plastic waste is through the process of recycling plastic materials to create novel products [2,3]. In this manner, the implementation of this approach can effectively contribute to the mitigation of waste accumulation in landfills, the preservation of valuable resources, and the safeguarding of the environment against the adverse impacts of plastic pollution, greenhouse gas emissions, and the disposal of plastic waste [4,5]. The production of plastic has been in existence since the 20th century. According to available data up until 2015, the total amount of plastic garbage recorded was 6.3 billion tonnes. However, only a mere 9% of this material was subjected to recycling, and a mere 1% underwent multiple recycling processes [6].

The plastics that are frequently subjected to recycling processes in Malaysia include polyethylene terephthalate (PET), high-density polyethylene (HDPE), and polypropylene (PP) [7]. The emergence of the post-consumer PET recycling business can be attributed to the increasing environmental pressure to enhance waste management practices. One additional element that contributes to the growth of the PET recycling industry is the slow rate of natural degradation of PET products [8]. Typically, polyethylene terephthalate (PET) is classified as a non-biodegradable polymer due to the absence of any known creature capable of metabolizing its rather massive molecular structure. The degradation of PET necessitates the utilization of biologically-based methods, which are both time-consuming and expensive [8,9]. The implementation of recycling initiatives for polyethylene terephthalate (PET) can yield numerous advantages in relation to the concept of sustainability. The utilization of PET material can be effectively extended by recycling processes, enabling the production of novel goods. This approach has been shown to enhance the stability of PET materials, resulting in improved efficiency and effectiveness [9,10].

PET was selected as the subject of investigation due to its potential to contribute to the diversification of plastic bottle recycling outcomes. This pertained to facts regarding waste management, specifically focusing on plastic bottle disposal. According to Pauline Goh, the general manager of the Malaysian Recycling Alliance (Marea), as reported in the New Straits Times on November 29, 2021, there is a projected fourfold deterioration in the condition of a significant portion of Malaysia's waste by 2050. This deterioration is attributed to the continuous increase in the domestic utilization of plastic packaging, highlighting the urgent need for collective and decisive measures to safeguard the environment. Based on available data, it is evident that Malaysia's general trash recycling rate remains quite low, standing around 30% [11]. However, it is possible that the amount of plastic waste generated from packing might be significantly reduced. It is imperative to develop a range of solutions and novel inventions centered around waste management. This study aims to further investigate polyethylene terephthalate (PET) in order to mitigate the wastage of used plastic bottles by exploring their potential for repurposing into new goods.

The demand for reprocessed and recycled products has been on the rise due to growing environmental consciousness among individuals. The examination of plastic bottles in this study can be associated with the concept of sustainability, as recycling played a crucial role in promoting sustainability by mitigating the disposal of recyclable materials in landfills. It contributes to the mitigation of environmental pollution. Plastics, in contrast to food and paper waste, exhibit a limited capacity for total elimination from the environment when subjected to decomposition in landfills [12]. The disposal of these items within the solid waste management cycle is a significant challenge. Plastics referred to as microplastics when their length is less than 5 mm, undergo a degradation process that spans several centuries to millennia, resulting in the formation of smaller plastic fragments [12-14]. As a result, the accumulation of plastics on Earth has led to a reduction in available landfill space, while also causing significant and nearly impossible to eliminate contamination of the natural environment [15,16].

The objective of this study was to determine the optimal combination of additive and polymer to be blended with recycled polyethylene terephthalate (PET). Methylene diphenyl diisocyanate (MDI), polypropylene-graft-maleic anhydride (PP-g-MA), and ethylene-butyl acrylate-glycidyl methacrylate (E-BA-GMA) are illustrative instances of compatibilizers. Polymers also exhibit a diverse range of types, such as polycarbonate (PC), polylactic acid (PLA), and polypropylene (PP).

Hence, the objective of the research was to perform mechanical evaluations on the r-PET/polymer mix using a compatibilizer, encompassing tensile, flexural, and impact assessments. The polymer and compatibilizer selected for this study are PC and MDI, respectively. These novel composites have the potential to yield significant advantages in the production of innovative items, particularly when employing the 3D printing technique. The association between 3D printing and plastic waste pertains mostly to the use of a filament.

2. Methodology

2.1 Material

2.1.1 Recycled Polyethylene Terephthalate (r-PET)

Recycled poly (ethylene terephthalate) (r-PET) was obtained by the post-consumer drinking plastic bottles. The waste of PET was washed and shredded into a pellet. Figure 1 shows the type of PET plastic bottle that was used in this experiment.

Fig. 1. Polyethylene Terephthalate (PET) plastic bottle

2.1.2 Polycarbonate

The polycarbonate (PC) resin was acquired from Alba.os, a supplier located in Johor, Malaysia. The material exhibited a melt flow index (MFI) of 22g/10min when subjected to a temperature of 300°C and a load of 1.2 kg. Polycarbonates exist in granular form, characterized by a melting point of 230°C and a density ranging from 1.20 to 1.22 g/cm³.

2.2 Composition and Designation

The composition of each material which are r-PET, PC and MDI that being used were determined and recorded as Table 1 below.

Table 1
Composition of r-PET/PC/MDI blend

Compositions	Abbreviations	r-PET (wt%)	PC (wt%)	MDI (wt%)
1	r-PET/PC1	20	80	0.9
2	r-PET/PC2	40	60	0.9
3	r-PET/PC3	60	40	0.9
4	r-PET/PC4	80	20	0.9

2.3 Sample Preparation
2.3.1 Recycling process of PET

The collection of PET waste is facilitated by a systematic sorting technique. After the completion of the sorting step, the PET is further subjected to a washing process. During the course of this operation, the label, cap, and other components of the plastic bottle were detached. After the elimination of all materials except for polyethylene terephthalate (PET), the PET is subjected to a crushing process using a specialized equipment to transform it into small pellets. The shredded polyethylene terephthalate (PET) was subsequently subjected to a 15-minute rinsing process in a 2% sodium hydroxide (NaOH) solution, where the molar mass of NaOH is 40.0 grammes per mole (g/mol). Following the washing process, PET flakes underwent a single rinse in tap water in order to eliminate any remaining NaOH and contaminants. Subsequently, polyethylene terephthalate (PET) was subjected to a drying process for a duration of 6 hours at a temperature of 75°C in an oven. Figure 2(a) depicts the shredded polyethylene terephthalate (PET) material subsequent to undergoing a thorough washing and rinsing procedure. On the other hand, Figure 2(b) illustrates the drying phase of the PET material, employing an oven for this purpose.

(a) (b)

Fig. 2. (a) The shredded PET after the washing and rinsing (b) drying process of the shredded PET

2.4 Blending and Mixing Process of r-PET/PC/MDI Composite

The blending process of r-PET and PC in the form of flakes was carried out by melting the material using a Brabender plastograph with the addition of MDI as the compatibilizer. The speed of the Brabender that is used was 30 rpm with a temperature profile of 230 °C.

2.5 Injection Molding

The r-PET, PC, and MDI composites were blended together and afterward pelletized. These pellets were then introduced into an injection moulding machine to manufacture specimens with predetermined properties. Following this, mechanical testing was conducted on the specimens. The temperature of the barrel in the injection molding process was set to 250 °C, while the mold temperature was maintained at 50 °C.

2.6 Mechanical Testing
2.6.1 Tensile test

The initial test conducted was a tensile test, following the ISO 527-2 specification. In order to obtain the average tensile result for each composition, a total of five specimens are evaluated. The test was conducted using the Instron 5567 Universal Testing Machine under ambient conditions, with a crosshead speed of 10 mm/min. An average value was obtained for each composition by evaluating five specimens, and this value was subsequently recorded.

2.6.2 Flexural test

The subsequent experimental procedure involved executing of the flexural test. The experiment was conducted in accordance with the ISO 178 standards, with an Instron 5567 universal testing machine. A crosshead speed of 5 mm/min is being used in conjunction with a support span of 100 mm under ambient conditions. The average values of flexural strength and flexural modulus were obtained by analyzing the findings of five specimens for each composition.

2.6.3 Impact test

The last examination was conducting the Charpy impact test, a widely accepted method for evaluating the energy absorption capacity of different compositions of r-PET/PC/MDI under high strain rates during fracture. The experiment involved the application of a test to assess the impact toughness of each composition, following the ISO 179 standard, using a hammer with a capacity of 4 J.

2.7 Data Collection

Data collection was conducted on all three experiments such as the tensile strength, Young Modulus, flexural strength, flexural modulus and the impact strength.

2.7.1 Young Modulus

Young's Modulus, denoted as E and also referred to as the tensile modulus or modulus of elasticity, is a fundamental mechanical property that quantifies the stiffness of a solid material under tensile or compressive forces applied along its length [17]. The subsequent equation is employed to calculate the Young Modulus, denoted as E:

$$Young\ Modulus, E = \frac{Stress}{Strain} \tag{1}$$

2.7.2 Flexural strength and flexural modulus

The flexural strength and flexural modulus of the test were determined by the utilization of the following equations respectively.

$$Flexural\ Strength = \frac{3FL}{2bd^2} \tag{2}$$

$$Flexural\ Modulus = \frac{l^3 \Delta W}{4bd^3 \Delta S} \tag{3}$$

where *L*: Span between the centers of support (m), *F:* Force (N), *l*: Length of sample (m), *B*: Width of the sample (m), *D:* Thickness of the sample (m), *ΔW*: Increment in load (N), *ΔS*: Increment in defection

3. Results
3.1 Tensile Strength and Young's Modulus

The outcomes of the tensile tests conducted on four distinct ratios are presented in Figure 3 below. The r-PET/PC1 composite exhibited the maximum tensile strength, measuring 22.30 MPa. The tensile strength of 22.01 MPa was observed to be the second highest among the four ratios, suggesting that this value corresponds to the r-PET/PC2 ratio. In comparison to the reported value of 0.30 MPa for r-PET/PC1, a little decrease was observed in the case of r-PET/PC2. The tensile strength of the r-PET/PC3 material was determined to be 21.21 MPa. The final recorded value for r-PET/PC4 was determined to be 17.95 MPa. When comparing r-PET/PC4 to the other three ratios that were examined, it was found to have the lowest value.

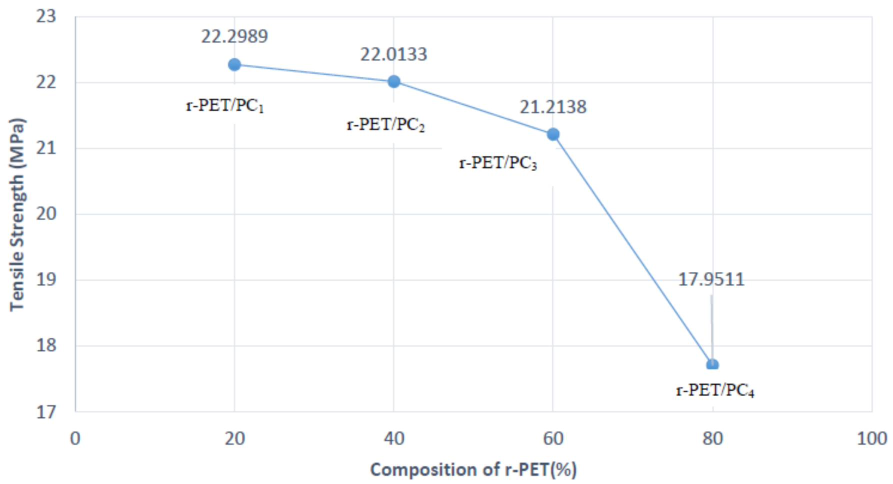

Fig. 3. Graph of Tensile Strength of r-PET/PC/MDI blend

The experimental findings indicate that a comparison between recycled PET polymer and virgin PET is not feasible due to the superior mechanical capabilities and thermal stability exhibited by the latter [18]. The average tensile strength decreases as the percentage of recycled polyethylene terephthalate (r-PET) in the mixture increases, as seen by the mean value obtained from the tensile test. The average tensile strength of the r-PET/PC1 composition suggests that it possesses superior tensile resistance compared to other ratios being evaluated. In contrast, r-PET/PC4 exhibits the lowest tensile strength, indicating its classification as a composition with low tensile capacity.

The graphical representation of the observed impact under evaluation in this experiment was illustrated in Figure 4. The graph below illustrates a positive correlation between Young's modulus and r-PET content. The Young's modulus of the r-PET/PC1 composite material was determined to be 1.88 MPa, indicating that it exhibited the lowest modulus value among the other tested ratios. The r-PET/PC4 material exhibited the greatest Young's modulus value, measuring 2.95 MPa. The data provided includes the r-PET/PC2 and r-PET/PC3 values, which were measured to be 2.07 MPa and 2.28 MPa, respectively.

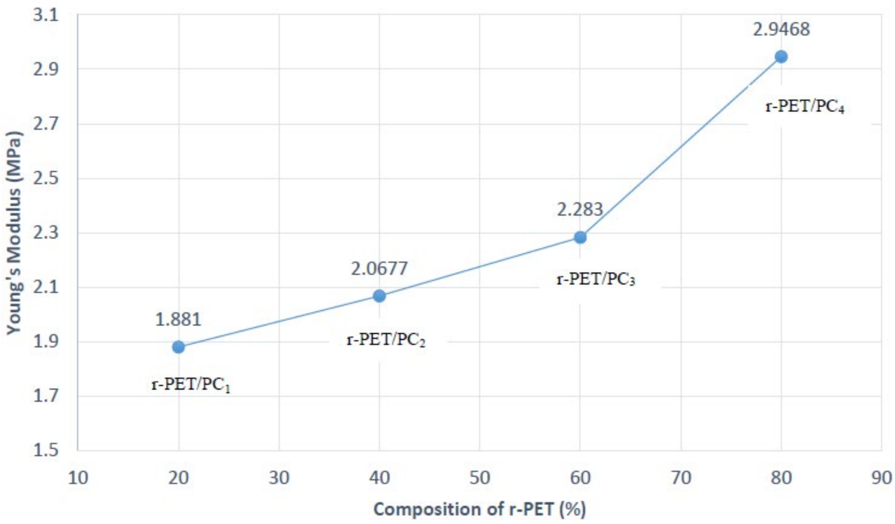

Fig. 4. Graph of Young's modulus of r-PET/PC/MDI blend

The Young's modulus of a rigid material is characterised by a high value, whereas the Young's modulus of a pliable material is characterised by a low value. The results obtained for the r-PET/PC4 in this particular case study illustrates the characteristics of a more rigid material in comparison to the other ratios. During the test, it has been shown that r-PET/PC4 specimens exhibit a tendency to fracture at a distinct load point. The proportion of r-PET in the mixture exhibited the highest value among the remaining components. This statement aligns with the notion that the stiffness of a component is indicative of its ability to undergo deflection when subjected to a specific load [18]. The r-PET/PC1 material exhibits elastic composition behaviour, wherein the specimen has a tendency to undergo elongation during the testing process [19]. The r-PET content in the blend exhibited the lowest value when compared to the other ratios examined. Other than that, this pattern of result are also observed by previous researcher that indicate that transesterification between PET and PC along with the chain extension introducing by MDI produced copolymers and polymer network that had good interfacial adhesion between phases resulting on the new polymeric system [20].

3.2 Flexural Strength and Flexural Modulus

The data obtained during the course of the experiment is presented in Figure 5. As previously mentioned, five specimens were assessed for each ratio. The average flexural strength value is calculated by utilizing five individual values. Subsequently, a comparison is made between the numbers. In this experiment, it was observed that the r-PET/PC2 material had the highest average flexural strength, measuring 121.41 MPa. The minimum recorded value was 68.44 MPa, specifically attributed to the r-PET/PC4 material. The flexural strength values of r-PET/PC1 and r-PET/PC3 were found to have average values of 119.77 MPa and 109.61 MPa, respectively, with the former being the highest value and the latter representing the lowest value.

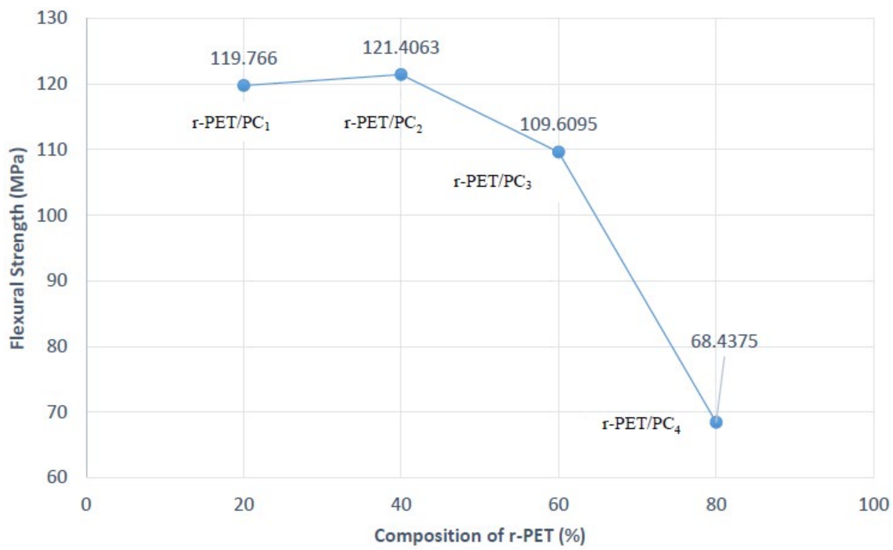

Fig. 5. Graph of average flexural strength r-PET/PC/MDI blend

In the context of a uniform substance, the flexural strength is equivalent to the tensile strength. In the majority of the examples examined, the flexural strength of the material was found to be greater than its tensile strength [21]. The empirical data obtained in this investigation substantiated the aforementioned assertion. The observed average flexural strength value was found to be higher than the observed average tensile strength value. Flexural strength refers to the capacity of a specimen to withstand bending deflection under the application of energy to the structure [22]. The results presented indicates that among the four ratios examined, r-PET/PC2 exhibits the highest resistance to bending forces. In contrast, r-PET/PC4 demonstrates a limited capacity to withstand bending force.

The graph in Figure 6 illustrates the flexural modulus for each ratio being examined. The flexural modulus formula was employed to compute all of the values. The graph exhibited a state of variability. The initial ratio, denoted as r-PET/PC1, was found to be associated with a flexural modulus value of 56.26 MPa. The flexural modulus value of r-PET/PC2 was 62.64 MPa, which was the second highest among the materials tested. In comparison, r-PET/PC3 had a flexural modulus value of 58.48 MPa. Among the four ratios examined, it was observed that the r-PET/PC4 ratio exhibited the greatest flexural modulus value, measuring at 92.27 MPa. A notable disparity was seen between the maximum and minimum values, both of which measured 36.01 MPa. This phenomenon is commonly referred to as the disparity in their mechanical response with respect to bending force.

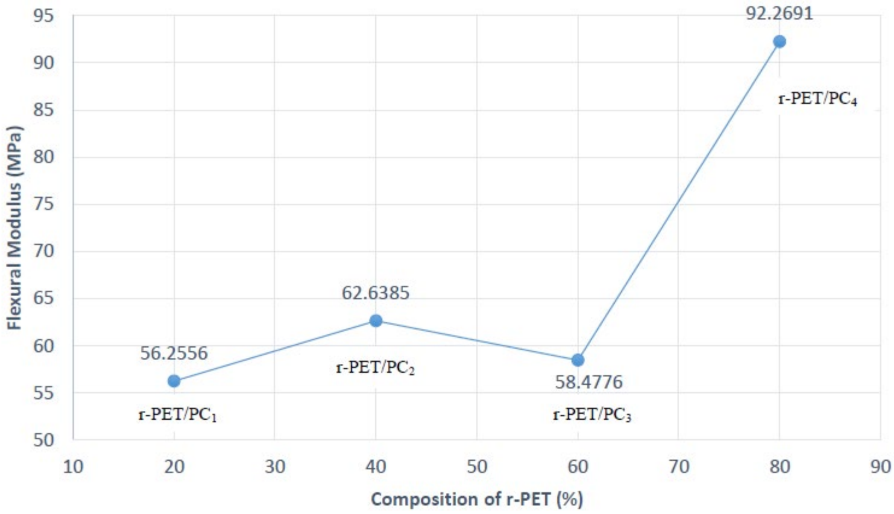

Fig. 6. Graph of flexural modulus of r-PET/PC/MDI blend

A correlation was seen between the flexural modulus of the material and its resistance to bending. According to previous researcher, an increase in the flexural modulus of a material is directly associated with an increase in its resistance to bending [23]. The data displayed in the graph serves as a representation of the relationship under discussion, as it has been calculated and stored for analysis. When considering the various ratios, it can be observed that the r-PET/PC4 composition exhibited the highest level of difficulty in terms of its performance in bending testing. On the contrary, r-PET/PC1, being the lowest flexural modulus, facilitates the bending of the composition. The value of r-PET/PC3 is somewhat higher than that of r-PET/PC1, and its mechanical characteristics differ from r-PET/PC1 in that it exhibits greater flexibility while being able to resist larger bending deflections. In contrast, the r-PET/PC2 ratio exhibited a notable increase in flexural strength but was accompanied by a comparatively lower flexural modulus in comparison to the alternative ratio under investigation. The composition provides an account of the motion shown by the specimen under conditions of significant bending deflection, while also highlighting its inherent ease of bending.

3.3 Impact Strength

The resulting impact strength value from the test is depicted in Figure 7 below. The highest recorded impact strength obtained in the study was 0.32 J, specifically associated with the r-PET/PC1 material. The r-PET/PC2 material achieved a value of 0.23 J, placing it in the second position in terms of impact strength. The reported values for r-PET/PC3 and r-PET/PC4 were 0.14 J and 0.09 J, respectively.

The provided graph illustrates the relationship between the impact strength and the r-PET content in the blend. As the proportion of r-PET in the blend increased, there was a corresponding decrease in impact strength. The following information elucidates the behavioural patterns exhibited by each of the ratios in the test. The composite material with the highest average impact strength consists of 20% r-PET and 80% PC, by weight. The Virgin PC exhibits superior mechanical properties and progressively boosts the impact strength of the material under examination for this particular ratio. The level of interaction among the molecules within the mix is directly proportional to the material's impact resistance. In contrast, the composition consisting of 80% r-PET and 20% PC by weight exhibited the minimum average impact strength value. In this composition, a reduced amount of virgin polycarbonate (PC) was utilized, while a greater proportion of recycled polyethylene terephthalate (PET) was applied. The reduced impact strength of the composite was attributed to the inferior mechanical properties of r-PET [24].

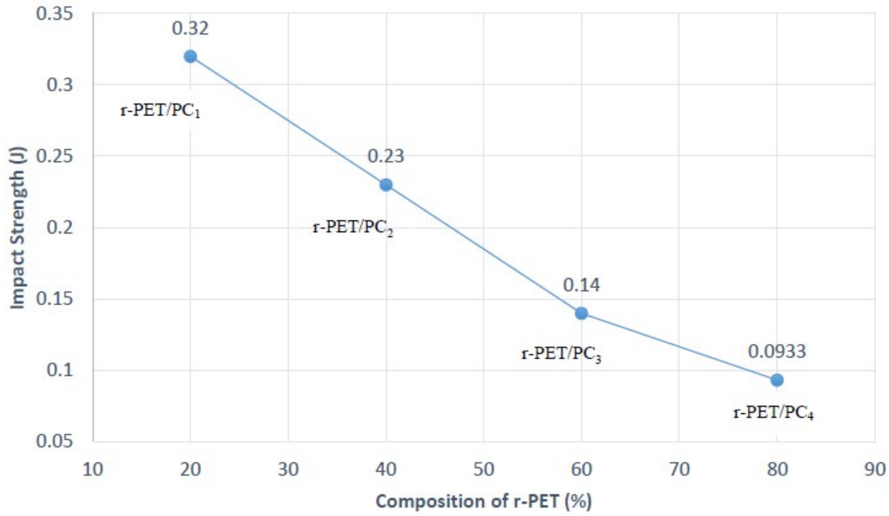

Fig. 7. Graph of average impact strength of r-PET/PC/MDI blend

4. Conclusions

Polymers are frequently utilised in modern day society across a wide range of applications, both in consumer goods and industrial processes. The objective of this study was to utilize recycled polyethylene terephthalate (PET), commonly sourced in the form of mineral plastic bottles procured from various manufacturers. The mechanical properties of recycled PET were found to be inferior to those of virgin PET. The utilization of MDI for enhancing the mechanical properties of the r-PET/PC blend yields a favorable conclusion, as the addition of PC to the r-PET mixture effectively enhances its mechanical features.

In order to enhance the mechanical properties of the mix, four distinct weight percentages were employed in varying ratios of r-PET/PC, specifically 20/80, 40/60, 60/40, and 80/20. The MDI composition remained consistent at 0.9% for all four ratios. Furthermore, by the consideration of the outcomes obtained from the conducted experiment, it was feasible to deduce the subsequent findings:

i. According to the recorded data on tensile strength, the ratio of r-PET/PC1, consisting of 20% r-PET and 80% PC by weight, exhibited the highest value among all the ratios. Conversely, the ratio of r-PET/PC4, also composed of 20% r-PET and 80% PC by weight, demonstrated the lowest value. In contrast, Young's modulus exhibited contrasting results, with r-PET/PC1 demonstrating the lowest value and r-PET/PC4 exhibiting the highest value. Based on the available data, it can be observed that the r-PET/PC1 combination has characteristics akin to a material capable of withstanding a greater load prior to buckling. However, it demonstrates a deficiency in stiffness throughout the elastic region. The contrary behaviour of r-PET/PC4 was observed, wherein the material exhibited increased load-bearing capacity but also had a larger stiffness factor inside the elastic zone. The incorporation of PC content into the mix resulted in a decrease in Young's modulus, while all four ratios exhibited a consistent rise in tensile strength.

ii. The findings pertaining to the flexural strength and flexural modulus exhibited dissimilarities, as the flexural modulus demonstrated an increase while the flexural strength exhibited a decrease. The flexural strength of the composite material consisting of 40% r-PET and 60% PC, known as r-PET/PC2, exhibited the highest recorded value among all tested specimens. Nevertheless, the aforementioned ratio just documented the second-highest value of flexural modulus, rather than the lowest. The material denoted as r-PET/PC4 exhibited the lowest flexural strength, while simultaneously possessing the highest flexural modulus value. The composition that exhibited the greatest resistance to bending deflection under the application of energy, but with a somewhat reduced propensity for bending, was r-PET/PC2. The r-PET/PC4 material has a pronounced inclination towards bending, although displays little resistance against bending deflection. The improved bending capabilities of r-PET/PC2 can be attributed to the strong bonding between its constituent structures, which ultimately leads to the formation of a stronger material.
iii. An analysis was conducted on the acquired data pertaining to impact strength. The impact strength value of r-PET/PC1 was found to be the highest, whereas r-PET/PC4 exhibited the lowest impact strength value. As the proportion of r-PET in the mix was increased, a steady decrease in impact strength was seen. The highest concentration of polycarbonate (PC) was observed in the r-PET/PC1 composite, resulting in increased molecular interaction between r-PET and PC. Hence, the material exhibits enhanced resistance to impact when subjected to a sudden application of load. However, because of the limited mechanical properties of r-PET, increasing the proportion of r-PET in the blend leads to a decrease in the material's strength.
iv. The findings indicate that achieving high mechanical properties in the r-PET/PC/MDI mix can be accomplished by the customisation of the PC component's weight percentage. In this particular study, it was determined that the most effective ratio for enhancing mechanical properties was the use of r-PET/PC1. Furthermore, other than exhibiting exceptional tensile and flexural strengths, it also demonstrates commendable performance in terms of impact strength.

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