Rheological and Mechanical Investigation of PET, LDPE and HDPE Wastes Blended for Re-Use in Bio-Pipes

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Abstract. This work focuses on the re-use of biopolymer wastes to produce the pipes and reduce the impact of these materials on the environment. The ratios of 10, 20, 30 and 40wt% of recycled polyethylene terephthalate (RPET) were added to the reference blend, which consists of recycled low-density polyethylene (RLDPE) and recycled high-density polyethylene (RHDPE). Rheological and mechanical tests were performed on these blends. The blend of RLDPE and RHDPE was already successful in the manufacturing of pipes. The capillary rheometer was used to check the shear viscosity and shear stress behaviour with the shear rate increasing for different blends. The density, tensile strength, elastic modulus and impact strength were also tested for all blends. The results showed that the shear viscosity decreases and the shear stress increases with the shear rate increasing for all blends. In general, the addition of RPET to the reference blend decreases the viscosity at each shear rate. The blends of 10 wt% and 20wt% are more compatible with the reference blend, while the blends of 30wt% and 40wt% exhibit a clear deviation after a shear rate of 300. The density indicates an increase with the increase of RPET up to 30%. After that, the tensile strength decreases and impact strength increases up to the 40wt% RPET. The results showed that the rheological test can be used to predict the mechanical behaviour. Additionally, there was a good agreement observed between the rheological and mechanical tests. The ten wt% and 20wt% blends were more suitable for this task.

Keywords. Polymers Wastes, Rheological Behaviour, Mechanical Tests, Bio Pipes.

1. Introduction
Plastics are synthetic polymers with a high resistance to acids, bases and chemicals. Plastics are resistant to microorganisms; therefore, they are not biodegradable. One plastic that is extensively used in certain industries, such as textile and packing, is polyethylene terephthalate (PET) [1]. PET is a linear polyester [2]. PET's important applications in packaging include the manufacturing of soft drink bottles and other beverages and fibre production.

PET has many important features, including high compressive and tensile strength, good chemical resistance, high impact resistance, transparency, fracture resistance, gas penetration resistance, heat resistance, good electrical properties, low weight and a lower price compared to other packaging materials, such as glass and metals. PET has a long life due to its resistance to biodegradation, which leads to the accumulation of large amounts of PET waste. Due to the development of new technologies, it is
impossible to avoid the use of plastics. Therefore, plastic materials should be recycled [1]. Recycling is the main process to reduce waste material [3]. The following two important techniques are used to recycle waste PET: chemical recycling and mechanical recycling. Mechanical recycling of PET waste typically includes the elimination of contamination by separating, washing, drying and melting [1]. Mechanical recycling is the most desirable approach because of its low cost and high reliability. In general, mechanical recycling keeps the molecular structure of the polymer molecule intact [4]. A mechanical recycling method that involves blending the plastics is one of the solutions for recycling plastics [5]. However, exposure to several rounds of reprocessing recycling may change the molecular structure and molecular weight of PET, which changes the rheological and mechanical properties of PET. Therefore, a mixture of PET with other polymers, in particular polyolefin, may produce a significant equilibrium in the mechanical properties and processability [1].

High-density polyethylene (HDPE) and PET have been widely used in packaging applications and constitute a large portion of post-consumer wastes [5]. HDPE is mostly linear and much more crystalline [6]. It is one of the largest used commodities, thermoplastic, for industrial and household applications. Its mechanical properties make it an ideal material for moulding products, and it is a 100% recyclable material [7]. Low-density polyethylene (LDPE) is a highly branched form [8] and a highly flexible material because it contains numerous side chains that increase the distance between the main C-C chains [9]. LDPE contains short-chain propyl- and butyl branches, as well as a few long-chain branches. Table 1 illustrates the structures of these recycled polymers [8].

| Names                  | Structure                       |
|------------------------|---------------------------------|
| Polyethylene (PE)      | [CH₂-CH₂]n                      |
| Polyethylene terephthalate (PET) | [CCCH₂O]₀       |

Rheology is a branch of material science that deals with the deformation and stream of matter under stress [10]. In academic studies, rheology science is a significant field, and it affects the plastics industry, sprays, detergents, medical research, food and others [11]. Rheological and physical properties are used as an indication as to whether the recycling process is going in the right direction [12]. The elastic modulus is a measure of the stiffness of the material [13].

A capillary rheometer is commonly used in both industries and academia to determine the rheological behaviour of polymer melts at high shear rate levels before evaluating their processability on a full industrial scale [14]. The capillary rheometer is the most frequently used method for determining the flow properties of polymer melts. The fluid property results provided in the capillary rheometer depend on the device's design, with the same die giving different results when used in different device models [15].

2. Materials and Methods

Recycled low-density polyethylene (RLDPE) from agricultural covers waste and recycled high-density polyethylene (RHDPE) from fruits boxes and chairs waste were provided from the Al Ghadeer factory in Karbala in Iraq as pellets. The RLDPE and RHDPE blend is a reference blend that was previously used to produce pipes, as seen in Figure 1. The recycled polyethylene terephthalate (RPET) that was used in this
study was collected from water bottle waste from the Iraqi market. After crushing, washing, drying and converting the wastes into flakes, the properties of these recycled polymers are shown in Table 2.

![Figure 1](image.png)

**Figure 1.** The pipes manufactured from the RLDPE / RHDPE blend.

The outer and inner diameters are 31.9 mm and 21.7 mm, respectively.

**Table 2.** Properties of the used RPET, RLDPE and RHDPE.

| Properties            | RPET  | RLDPE (Branch) | RHDPE (Linear) |
|-----------------------|-------|----------------|-----------------|
| MFR (g / 10min)       | 0.72  | 9.60           | 5.34            |
| Density (g/cm³)       | 1.1584| 0.8696         | 0.9066          |
| Melting point (°C)    | 250   | 115            | 137             |

3. **RPET**

The waste from the old plastic’s water was collected from multiple sources, the dirt and stickers were washed away and it was dried. Then, the old plastics were cut, which is usually done to reduce the initial volume. The process is conducted using scissors to cut it into longitudinal slices and then small pieces. Then, the cutting machine is used, which contains a series of rotary blades that are driven by an electric motor. The small pieces of PET are placed in the slicing machine via the hopper located above the rotor blade. The slicing product is a pile of plastic flakes, which are rough and irregularly shaped, and they can then be treated. Every 50 old plastic pieces weigh 320 g.

4. **Sample Preparation**

The polymer blend of RLDPE, RHDPE and RPET are extruded using a co-rotating twin-screw extruder model (SIJ-30A). The diameter of the screw is 30 mm, and the screw speed is zero–320 rpm. The polymer blends are prepared in the following procedures: RLDPE with 62, 55, 49, 43 and 37 wt%, RHDPE with 38, 35, 31, 27 and 23 wt% and RPET with ten, 20, 30 and 40 wt%. These polymers were mixed at room temperature according to the ratios displayed in Table 3 to produce sheets for testing.
Table 3. Blends ratios that were used in this work.

| Sample NO. | Constituent |     |     |     |
|------------|-------------|-----|-----|-----|
|            | RPET (wt%)  | RLDPE (wt%) | RHDPE (wt%) |
| 1          | 0           | 62   | 38  |
| 2          | 10          | 55   | 35  |
| 3          | 20          | 49   | 31  |
| 4          | 30          | 43   | 27  |
| 5          | 40          | 37   | 23  |

The mixed materials were placed in a co-rotating twin-screw extruder at a constant temperature (210°C) and constant speed (30 rpm) until the resulting sheets were ready to be used in mechanical tests.

5. Characterisation

5.1. Rheological Measurement
The rheological behaviours of different (RLDPE/RHDPE/RPET) blends were determined using a capillary rheometer (SR20) (INSTRON CEAST, Country of Origin Italy) that was equipped with a capillary length of 20 mm and a diameter of one mm. The sample weights were approximately ten g with the shear rate in the range of 100–500 s⁻¹ at a temperature of 250°C. The samples were added to the capillary, heated to a specific temperature, held for ten minutes, and then tested at different stresses on their rheological parts. All rheological measurements were performed in rheological parameters and were collected directly from the supplier’s computer software (TRIOS, TA Instruments, New Castle, DE, USA).

5.2. Density Test
According to the ASTM D-792, density is measured at room temperature via a digital accuracy of ± 0.0001 g/cm³ using Matsu Haku High Precision DENSITY TESTER GP-120S. This method can be used with a sheet, rod, tube, moulded articles and powders. The specimen was weighed in both the air and distilled water at 23°C using a sinker and wire to hold the specimen completely submerged as required.

5.3. Mechanical Characterisation

5.3.1. Stiffness of the Pipe
Pipe stiffness is an inherent strength that is measured according to the standard ASTM D2665 test method for the determination of external loading characteristics of plastic pipes. The pipe is deflected at five per cent to the internal diameter, and the load is recorded.
5.3.2. Tensile Test
Instron tester type 5556 Universal Testing Machine (WDW/5E) in accordance with ASTM D638-IV was used to test the tension properties at a crosshead speed of five mm / min. The elastic modulus was also calculated depending on the data from the tensile test.

5.3.3. Impact Strength
The Charpy notched impact samples, according to the ASTM D-256, with dimensions of 65×13×3.2 Pmm³ were tested by a cast pendulum impact tester. A blunt notch with a notch tip radius of 0.25 mm was introduced to the impact specimens with a CEAST notch opener.

6. Results and Discussion

6.1. Rheological Properties

6.1.1. Viscosity Curve
The shear viscosity behaviour curve shows significant indications about the structure changes of different blends, as shown in Figure 2. Most polymeric materials in the viscosity curve show shear-thinning behaviour, and the polymers used in this research are among them. The shear behaviour indicates changes that occur in the structure and chains of the polymer during the extrusion process. These changes are related to the rheological behaviour during the extrusion process, as well as to the final specifications of the polymer hardened after extrusion. Therefore, it is important to identify the shear viscosity behaviour of the blends and study the changes that take place.

![Figure 2. Shear viscosity behaviour vs. shear rate of different LDPE/HDPE/PET recycled blends.](image)

In Figure 2, the shear viscosity decreases with the increase of the shear rate of the first blend (RLDPE and RHDPE). This blend is to prove success in the manufacturing of the pipe. The viscosity decreases rapidly from 100 to 200 shear rates, and then gradually slopes to 300 shear rates. Finally, the viscosity becomes more stable with the shear rate of up to 500. After adding the RPET to the first blend by ten wt%, the same style of shear-thinning behaviour is observed with an almost equal decrease along the curve by 100pa.s. When adding 20wt% of RPET to the first blend, the resulting viscosity curve is less than the viscosity of the first blend by approximately 60pa.s. However, when adding 30wt% of RPET to the first blend, the viscosity behaviour is similar to the previous three blends up to the shear rate of 300. After that, the
viscosity deviates from the usual behaviour. Finally, when adding 40wt% of the RPET, the shear viscosity decreases rapidly until the 300 shear rate. Then, the speed continues to decrease with less intensity but more values compared to the rest of the ratios.

According to the foregoing, the closest blend to the first blend with the rheological behaviour is the blend with a ratio of 20wt%, followed by the blend with a ratio of ten wt%. The last blend with a ratio of 30wt% and 40wt% is unstable and differs to the behaviour of the first blend [16].

6.1.2. Flow Curve

Figure 3 shows that the shear stress gradually increases with the shear rate increase of the first blend until the shear rate is 300. Then, it rises faster and moves towards stability.

![Figure 3. Shear stress behaviour vs shear rate of different LDPE/HDPE/PET recycled blends.](image)

When adding ten and 20wt% RPET to the first blend, the shear stress of these two blends behaves harmoniously and similar to the first blend. Note that the increase in stress is gradual from 100 to 400 shear rates, and then the trend leans towards stability for both blends. Shear stress increases gradually in the 30wt% RPET and 40wt% RPET blends until the shear rate is 400. This oscillation and diffraction that occurs from the natural pathway reflects the change in the composition of the polymers during movement and is consistent with the behaviour of the shear viscosity of the same blends in Figure 2. In general, the ten wt% and 20wt% blends are both closer to the first blend until the shear rate is 300. Therefore, they are more suitable for the aim of this work.

6.2. Density Test

Figure 4 indicates an increase in the density of all mixtures by adding different percentages of RPET because its density is greater than the other two materials. The density values shown in this figure are reasonable based on the densities of the three substances in Table 2, as the highest density is 1.07(g/cm³).
Figure 4. Density of the RPET / RLDPE/ RHDPE blends as a function of the RPET concentration.

The crystallinity in polymeric materials is directly proportional to the density and inversely proportional to the branching of its chains. This means that an increase in the percentage of crystallisation reduces the proportion of branching. This change in the composition of the mixtures directly affects the remaining mechanical, physical and thermal properties. Therefore, it is possible to predict what might happen with the two characteristics of tension and impact resistance. When they were examined, the first increased and the second decreased. From the foregoing, density testing can be used to either limit the most acceptable options or obtain initial indications about products [17].

6.3. Mechanical Properties

6.3.1. Stiffness of the Pipe
Pipe stiffness is a characteristic strength of the pipe. Pipe stiffness refers to the resistance to deflection. The greater the pipe’s wall thickness, the greater the stiffness, and therefore, its ability to resist external bending loads. The pipe manufactured from the RLDPE / RHDPE blend was tested, and the applied load and the amount of vertical deflection were 0.084 kN and 1.085 mm, respectively. The stiffness of the pipe ring was 5109.7 MPa and can resist cracks and return to its original position without permanent deformation. The stiffness of the manufactured pipes from the RLDPE / RHDPE blend was used as a reference to check the mechanical properties of the suggested blends. There is a direct relation between the tensile strength, Young’s modulus and stiffness. Increasing the tensile strength and Young’s modulus increases the stiffness, which is important for during pipe manufacturing. In general, the ten wt% and 20wt% blends are more acceptable regarding this property.

6.3.2. Tensile Strength
Figure 5 shows that the tensile strength increases in all mixtures with an increase in the threshold ratio of 30wt%, after which it tends to decrease. Its highest value is 35 MPa. This increase is due to the increase in the percentage of crystallisation and density and the decrease in the proportion of the branching chains in the compounds.
Figure 5. Tensile strength of the RPET / RLDPE/ RHDPE blends as a function of the RPET concentration. Therefore, these results are identical to the results of the density test, which is consistent with the previous analysis in Figure 4. The decrease in tensile strength at 40wt% is due to a change in the crystallisation of the chains’ branching ratio and the non-homogeneous distribution in this blend. The tested samples for tensile strength were produced at a certain shear rate. However, the viscosity and chains branching decrease and crystallinity increases with the RPET additions in each shear rate. The melting points of each polymer in Table 2 are another indication of the crystallinity content, as well as the structured content in the blends later [17,18].

6.3.3. Elastic Modulus (E)
In Figure 6, the modulus of the elasticity (tensile modulus) increases in all mixtures when increasing the RPET content, as it reaches its highest value of 100 MPa at 30wt% of the RPET content. This increase is attributed to an increase in the percentage of crystallinity, tensile strength, density and a decrease in the percentage of branching chains in the compounds. The elastic modulus decreased to 70 MPa for 40wt% RPET, which is higher than the value of the elasticity of the mixture before adding RPET.

Figure 6. Elastic modulus of the RPET / RLDPE/ RHDPE blends as a function of the RPET concentrations.

The elastic modulus is a measure of the stiffness of the material. Therefore, increasing the elastic modulus increases the stiffness of the material [13].
6.3.4. Impact Strength
One of the major mechanical tests where the material is subjected to a fast, dynamic load is the impact test. The impact test was conducted at room temperature on the samples using the Charpy method. Depending on the energy required for a sample fracture, the impact resistance was calculated for the polymer blend. Figure 7 shows the reference blend’s impact strength as a function of the RPET content.

![Impact Strength Graph](image)

**Figure 7.** The impact strength of the RPET / RLDPE/ RHDPE blends as a function of the RPET concentrations.

First, the impact strength decreases gradually and slightly at ten and 20wt% RPET, and it reaches its minimum value at 30wt% RPET. Then, it rapidly increases and reaches a maximum value of 40wt% RPET. This result matched the tensile, density and viscosity tests. The impact strength decreases with the viscosity, the and chains branching decreases at each shear rate.

From the above results, and according to the rheological and mechanical properties, it may be possible to manufacture similar pipes with better specifications. Ten wt% and 20wt% blends are more suitable for this task [19].

7. Conclusions
In the present work, PET waste (bottles and cans) was recycled and mixed with RLDPE and RHDPE in different ratios. The RLDPE and RHDPE blend that was produced was a successful pipe. The effect of the RPET content on the rheological and mechanical properties of the RLDPE / RHDPE blend was studied. The polymer blends exhibited shear-thinning behaviour. The shear viscosity decreased and shear stress increased as the shear rate increased for all blends. In general, the viscosity decreased as RPET was added at each shear rate. The rheological test can be used as a good indication of the mechanical properties of thermoplastic polymers, which reduces the cost, time and effort to trying to find the optimum results. There was an increase in the density and tensile strength and a decrease in the impact strength with the RPET added at a certain shear rate. The increase in the tensile strength and Young’s modulus indicates an increase in the stiffness, which is required for pipe manufacturing. The tensile strength and impact strength showed a good compatibility. Additionally, there was a good match between the rheological and mechanical behaviour.

In general, adding RPET to the reference blend decreases the viscosity at each shear rate. The decrease in the chains branching and density and the increase in the crystallinity is associated with the decrease in viscosity.
The increase of RPET to the reference blend up to 20wt% produced the same shear viscosity and shear stress behaviour with a lower value up to the shear rate 500. The blends of 30 and 40wt% RPET were compatible with the reference blend up to the 300 shear rate. After that, deviation occurred. According to the rheological and mechanical properties, similar pipes could be manufactured with better specifications. Ten wt% and 20wt% blends are more suitable for this task.

8. References

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