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Development and Evaluation of Recycled Polypropylene and Bean Pod Powder Composite Biomaterial for Fused Filament Fabrication

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Abstract

Approximately 450 million tons of plastic and agricultural waste are produced each year in the world. Only a small portion of this plastic waste is recycled, and a small portion of this agricultural waste is used as fuel or fertilizer, and the rest of this waste is left in the environment or is burned, resulting in environmental and air pollution. For proper disposal, plastic and agricultural waste can be used in the manufacture of composites as raw materials. In this study, we had evaluated the use of bean pod powder (BPp) was used as natural reinforcing filler in recycled polypropylene (rPP) based composites. BPp/rPP composite filaments were developed using the extrusion method and the samples were printed by Fused Filament Fabrication (FFF). Composites with rPP matrix containing different weight fractions of BPp (5%, 10% and 15%) were fabricated to observe and compare the mechanical properties (tensile, flexural, and compressive strength) of the filament composites. In addition, the filament surface was analyzed for roughness and particle size of bean pod powder. The results established that BPp/rPP composites exhibited better tensile, flexural, and compressive strength than rPP and pure PP. By adding 5 wt% BPp, the tensile strength of rPP increased from 20.4 MPa to 22.8 MPa. The highest flexural strength (15.05 MPa) was obtained at 5 wt% BPp among all composites and the highest compressive strength (24.5 MPa), was obtained at 10 wt% BPp. Therefore, it can be concluded that by carefully selecting the ratio of BPp to bean pod powder, it is therefore possible to positively influence the mechanical properties of the resulting composite.

Keywords

Pollution, Composite, Fused Filament Fabrication, Mechanical Properties

1. Introduction

Since their invention, plastics have become a key material in numerous manufacturing industries, including packaging, textiles, toys, sporting goods, electrical and electronic products, and agriculture [1]. In addition, plastics are also used in the fields of transportation, construction, and biomedical engineering [2]. Almost all consumer plastics are composed of hydrocarbons, and it is estimated that 8% of petroleum resources provide the materials and energy for modern plastics manufacturing [3]. Polymers are used to make consumer plastics (carbon-based molecules composed of repeating sequences of one or two basic units). To realize specific applications, polymers are selected based on their unique properties. Specifically, PET (polyethylene terephthalate) is a translucent plastic with good oxygen and moisture barrier properties, ideal for food packaging. Polypropylene (PP) and polyethylene (HDPE) are acid-and solvent-resistant plastics commonly used to package household soaps and detergents. The mechanical, performance, and durability of chlorinated polyethylene (PVC) are superior to those of other polymers, and their adaptability enables the production of flexible goods, packaging, and containers. When transparency is important, polystyrene (PS) can be used in rigid form or expanded for its insulating properties [4]. The main advantages of polymers are ease of processing, increased productivity and reduced costs. For many applications, reinforcing materials are used to optimize the properties of polymers to optimize mechanical properties [5]. Polypropylene (PP) is an interesting polymer for use in automotive parts. This is because polypropylene is a plastic suitable for additive manufacturing (AM) of custom parts, is inexpensive and has outstanding mechanical properties including high moldability, high durability, good chemical resistance, low density, and ease of processing. But this overuse of plastic is not without consequences. Polymers of fossil origin are one of the main sources of pollution, which also affects the consumption of oil during their production. Nearly half of the approximately 400 million tons of plastic waste produced annually worldwide is packaging [6]. Almost 60% of waste ends up in landfills and the environment, and at least 10% ends up in the ocean [7]. Most petroleum-based plastics are not biodegradable, and recycling only accounts for around 20% of the plastic produced globally [8]. Kenya generates approximately 1,334,000 tons of plastic waste each year, of which only 15% is recycled [9]. The government realized this challenge and put a ban on single use plastics. However, despite this intervention, the production and consumption of plastics keeps rising. Consequently, this continues to expose existing ecosystems to more pollution as inefficient systems such as landfills cannot keep up with the accumulation of plastic

[10]. Plastic take decades or centuries to break down naturally and do so via a process that produces greenhouse gas emissions that contribute to global warming. Plastic waste entering the marine food chain is often eaten by marine organisms [11]. While these effects are harmful to the environment and marine life, they are catastrophic for humans. Contact with nanoplastics and the harmful additives they contain can lead to health problems such as cancer, diabetes, obesity, and reproductive disorders, among others [7].

In addition to plastic waste, agriculture also produces a lot of by-products from harvesting and processing crops, causing environmental pollution. They damage the environment by releasing CO_2 during biodegradation [12]. Conventional methods of removing these crop residues, such as burning, increase damage by contributing to air pollution. In turn, landfill burial becomes a fire hazard due to decomposition problems [13] [14]. For disposal, most farmers choose to burn agricultural waste in their fields, which is the preferred method of disposing of pods in their fields because it is a quick, simple, and inexpensive process for farmers [15] [16]. However, the burning of agricultural waste results in severe soil and water pollution in the region. This practice significantly reduces the nutrients in the soil. For example, the complete combustion of atomic carbon, nitrogen, and sulphur results in the release of dangerous gases such as methane, nitrous oxide, and ammonia, which contribute to air pollution. In addition, these gases exacerbate already existing ozone pollution. Microscopic particles released during combustion are known to exacerbate chronic cardiovascular and respiratory diseases [15]. Sometimes the beans are sold to vendors without shelling, and after the vendors peel the beans, most of the pods are thrown into the public trash section, which contaminates the environment, and only a small portion thereof is fed to animals. In fact, bean pods are not the best fodder for livestock since their use sometimes causes animals to develop digestive complications (citation) get sick. Kenya is one of the largest producers of kidney beans in terms of distribution and the second largest producer after Uganda [17] [18]. Red beans, Mwitemania beans and broad beans are among the most widely produced pulses in Kenya. As can be seen in **Table 1**, the powders of the three main bean pods produced in Kenya after harvest were investigated. It was discovered that these powders include substances such Al_2O_3, S_iO_2, MgO, Fe_2O_3, and CaO, which are essential elements to improve the thermomechanical properties of composites.

After the review of plastic and agricultural wastes, the management of plastic and agricultural waste has been a major setback in environmental protection efforts. This is due to an unsustainable linear economic model based on a "get-make-dispose" structure that transforms raw materials into products that are eventually used and eventually discarded as waste in nature [19]. However, despite this model being dominant, finding novel ways to use plastic and agricultural waste as raw materials has advanced. Natural fibers are increasingly being used as reinforcement for plastics in place of fillers like glass, carbon, and minerals. In contrast to glass, carbon, and mineral fillers, this new class of materials uses natural fibers (wood, hemp, flax, etc.) as reinforcement for plastics. The development of biocomposites has been one of the most important developments in materials research over the past century [20]. Natural fibers are sustainable materials readily available in nature with the advantages of low cost, light weight, renewable, biodegradable, and high specific properties [19]. Cellulose, hemicellulose, and lignin make up the majority of the fibrous cell walls that make up natural plant fibers. A plentiful source of lignocellulosic materials with good mechanical properties is agricultural residues [21]. Because of their exceptional characteristics, which include enhanced mechanical strength, water and oxygen barrier properties, dimensional stability, heat, abrasion, and chemical resistance, low density, high availability, and low cost, natural fiber composites are becoming more and more significant [22]. Engineering, particularly mechanical, civil, and electrical engineering, has developed and used composite materials. Natural fibers are primarily used to produce materials with acceptable properties while lowering the cost of raw materials and environmental concerns [23]. Due to their environmentally friendly characteristics, natural or biocomposites are excellent alternatives to synthetic composites as environmental awareness grows. They also offer low cost, low density, flexibility, and reproducibility in comparison to or even outperforming their synthetic counterparts in these areas [24].

Fused Filament Fabrication (FFF) is the fastest growing of the additive manufacturing (AM) technologies and has great potential for biocomposite printing. FFF is widely used because of its many advantages, such as the ability to produce on a small scale, easy supply of raw material, limited amount of waste, high energy efficiency, and no expensive tools. It is nowadays used in industries and households [25]. In addition, the FFF process is less expensive and does not require harmful solvents or adhesives [26]. As shown in **Figure 1**, the FFF allows a convection of digital information into physical 3D objects [27]. The AM process begins with the design of a virtual 3D model using computer-aided design (CAD) software. Once the design is complete, the file is saved in STL format. FFF is seen as a technology that could potentially help recycle old plastic [28]. Its ability to print complex shaped objects that traditional manufacturers cannot produce and cheaper has attracted attention hence its use in several industrial fields [29] [30]. Furthermore, the printing device is small enough to be placed on a desktop [31]. In this technique, physical objects are obtained by depositing a molten material layer by layer along a predetermined path. Thermoplastic polymers and biocomposites are used in the form of filaments. Natural fibers have recently been widely used as additives in matrix polymers for filament fabrication [32].

However, finding the best combination of materials with different properties to meet engineering standards is a significant challenge. The fact that natural fibers are hydrophilic in nature poses compatibility challenges with plastic-based matrices which a mostly of hydrophobic in nature [5]. In addition, during filament

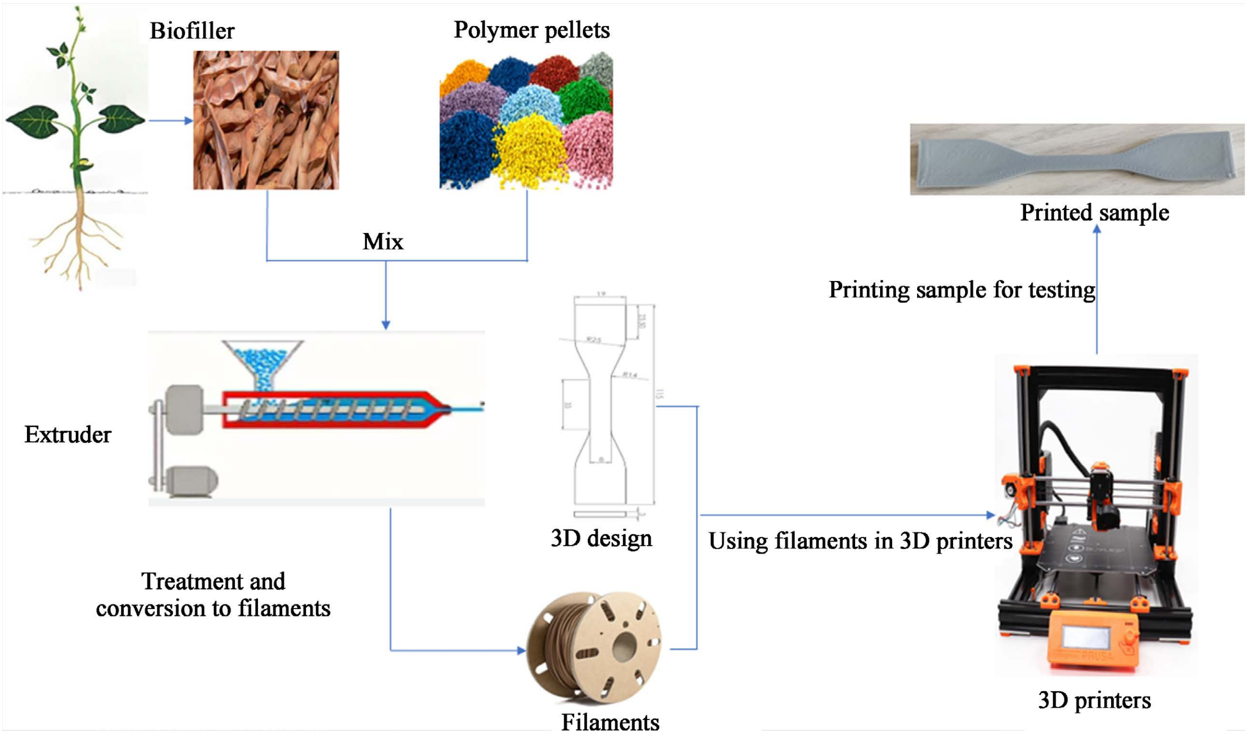


Figure 1. The cycle of FFF using natural fibers [34].

Table 1. Chemical composition of bean pods powder.

Constituents (%)	Red bean pods powder	Rosecoco bean pods powder	Green bean pods powder
SiO_2	38.19	27.45	11.73
Al_2O_3	4.61	2.03	3.18
MgO	9.36	9.97	8.67
P_2O_5	5.43	1.51	4.97
SO_3	2.91	1.77	3.59
K_2O	14.16	22.62	30.94
CaO	20.80	30.61	29.34
Fe_2O_3	3.46	2.96	5.16

making and during printing, natural fibers undergo thermomechanical degradation and the uneven distribution of fibers throughout the matrix is also a challenge that remains to be addressed [33]. Regarding the inclusion of bean pods in the AM process chain, as can be seen from **Table 1**, red bean pods show very good inclusion and research suitability. Indeed, apart from its good application potential, little attention has been paid to red bean pod-based biocomposites so far. This despite the fact that the physical and mechanical properties of biocomposites have been the subject of several studies. It is therefore necessary to study the effects of red bean pod powder on recycled polypropylene, which is the subject of this research work.

2. Materials and Methods

2.1. Materials

The recycled polypropylene pellets (rPP; diameter: 5.06 mm; width: 2.08 mm; color: white; humidity: 0.14%; melting point: 150˚C - 160˚C; melt flow index: 21 g/10 min at 230˚C, 5 kg) were purchased from Kenyaplastic (Nairobi, Kenya). Polypropylene filament "P-filament 721 grey" (diameter: 1.75 mm; color: grey; print temp.: 200˚C - 220˚C) were purchased from PPprint (Freiberg, Germany). The beans pod as an agricultural waste was collected from farmer base in Gatarakwa (Kieni, Nyeri County, Kenya).

2.2. Methods

2.2.1. Preparation of Beans Pod Powder

The bean pods were cleaned with water and dried in a MARUTO oven (model: S24A-2-333 serial number: 1994.0, Japanese) for 24 hours at 105˚C, as shown in Figure 2(a). The dried bean pods were ground by a grinder (XK06-002-00126, China) equipped with a 1 mm sieve. The bean pod particles obtained after grinding were placed in a set of sieves in descending order of fineness and agitated for 15 minutes, which is the recommended time for complete classification to obtain bean pods powder with a particle size of 75 μm, as shown in Figure 2(b).

The standard tests were performed for the characterization of bean pods powder and the results are presented in Table 2. The moisture was measured using the KERN DAB 200-2. The oxides composition of the BPp was obtained by X-ray fluorescence analysis using Bruker S1 TITAN 800 (China) and the result is showed in Table 1.

(a) (b)

Figure 2. (a) Bean pods after drying; (b) Bean pods powder.

Table 2. Physical properties of red bean pods powder.

Property	Value
Moisture content	0.90 (%)
Specific gravity	2.295
bulk density	1262 kg/m^3

2.2.2. Preparation of FFF Filaments

The extrusion technique was used to develop the composite filaments. The rPP pellets were shredded by 3 devo Filament Shredder (SHR3D IT) and dried at 75˚C for two (2) hours, the bean pods powder was also dried at 105˚C for twenty-four (24) hours in the oven to remove the moisture before mixing and extrusion. The rPP and bean pod powder were weighed and mixed in a sealed plastic box and then fed into an SJ35 single screw extruder (China). Filaments were made with proportions of 0, 5%, and 10% bean pods powder for melt blending with rPP. The recovered filaments were then cooled by passing through a water bath then continuously cut and shredded (**Figure 3**) then dried in the oven at 80˚C for two (2) hours to make sure it does not contain any moisture and finally re-extruded to obtain the BPp/rPP composite filaments (**Table 3**).

2.2.3. Preparation of Specimens

The 3D models of the specimens for tensile, flexural, and compressive, in accordance with ASTM standards, were computer modeled and exported as a

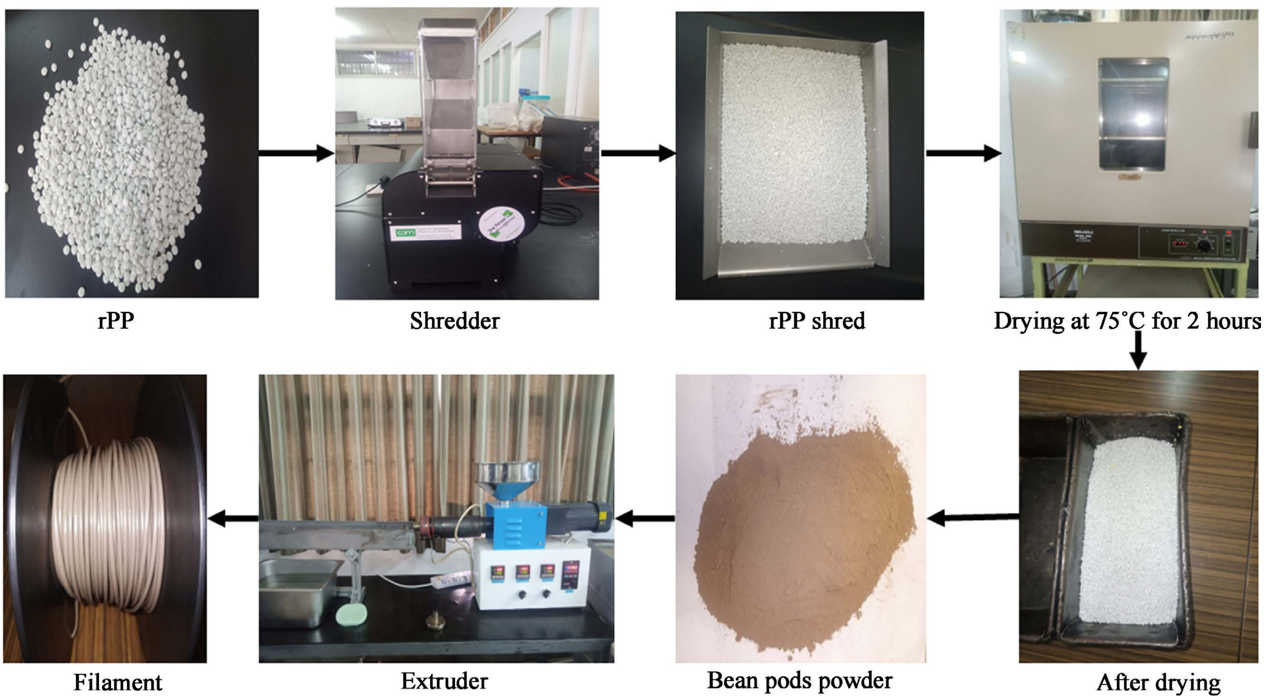


Figure 3. Filament production process.

Table 3. Load levels of reinforcing fillers for rPP and extruder parameters.

Composite filaments			Extruder	
Formulation	BPp	rPP	Temperature (˚C)	Speed (rp)
rPP	N/A	100	140 - 165	22
5-rPP	5	95	175 - 190	18
10-rPP	10	90	180 - 190	15.5

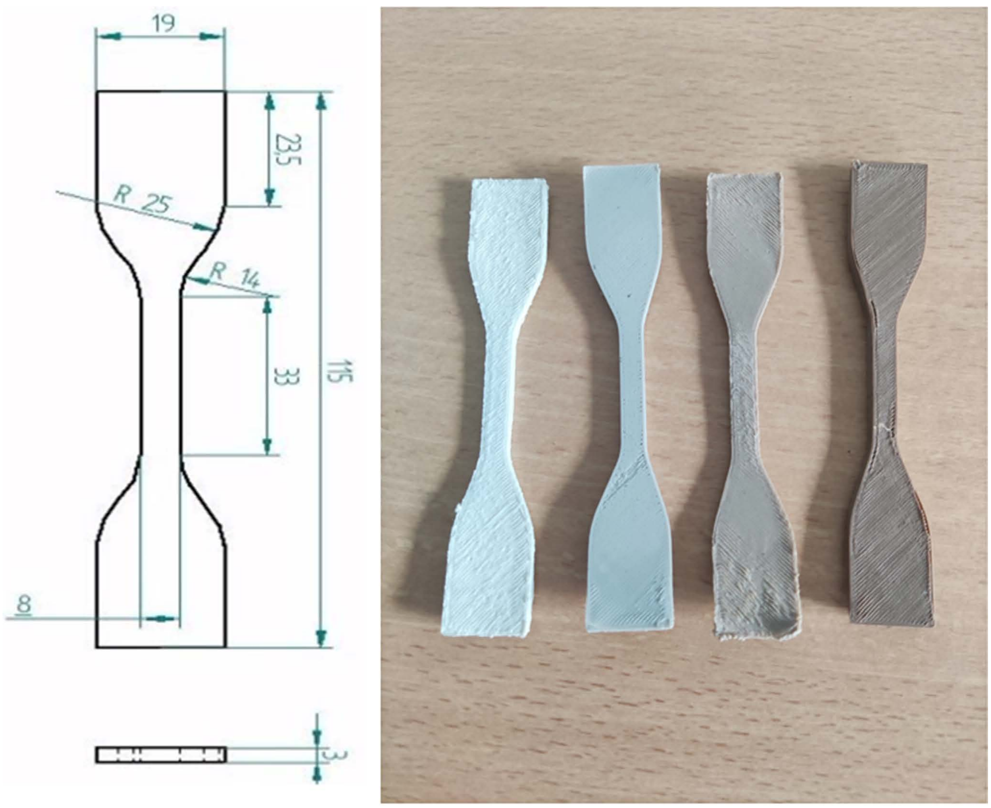
stereolithographic (STL) file, and then printed by a printer Prusa (Prusa i3 MK3) equipped with an MMU. Repetier-Server Pro 1.402-shiva-88 is used to control program parameters and control printers. The printing path of the layered sample is +45˚/−45˚, and the structural parts of the sample are made of filament segments with a printing width of 0.4 mm, a layer height of 0.1 mm, and a shell thickness of 1.2 mm. In addition, the printer nozzle temperature is 220˚C, the platform temperature is 80˚C for the first layer, 30˚C for the second layer, and the filling density is set to 100%.

Design (CAD) and printed part of each type of specimen are shown in the respective Figures 4-6.

2.3. Characterization

2.3.1. Surface Roughness Analysis

Rough surfaces typically have a peak-to-valley distribution. This is usually measured as the sum of negative and positive deviations from the "horizontal/central centerline" fitted to the surface of interest. Various surface amplitude roughness parameters are defined as: The arithmetic mean roughness Ra is the area between the roughness profile and its centerline. Rz is the difference between the highest "peak" and lowest "valley" of the surface. Ra and Rz are the most used parameters when measuring surface finish [35]. The surface roughness of filaments



rPP PP 5-rPP 10-rPP

(a) (b)

Figure 4. (a) ASTM D638 Type IV Tensile test 2D specimen, (b) Tensile test specimen printed.

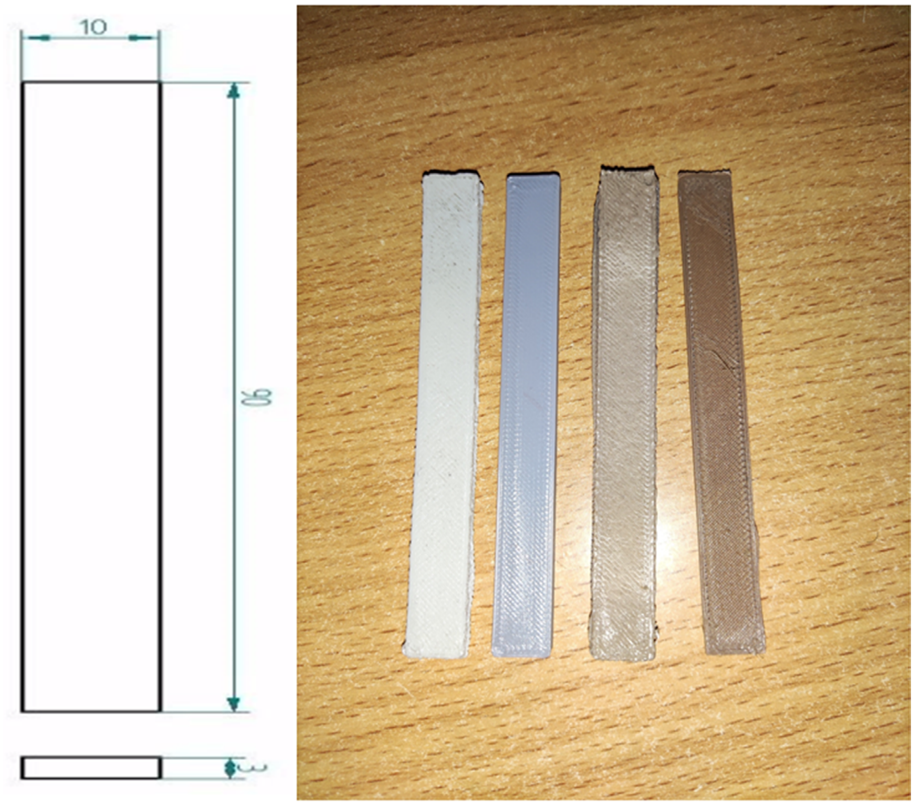


Figure 5. (a) ASTM D790 Flexural test 2D specimen, (b) Flexural test specimen printed.

Figure 6. (a) ASTM D695 Compression test 2D specimen, (b) Compression test specimen printed.

is measured according to the ISO 4287 standard using the MarSurf CM explorer, an accurate 3D surface measurement and analysis instrument.

2.3.2. Particle Size Analysis

The average particle size of the BPp was determined using a particle size analyzer (CAMSIZER XT, X-DRY, Retsch Technology), based on laser scattering.

2.4. Mechanical Tests

2.4.1. Tensile Test

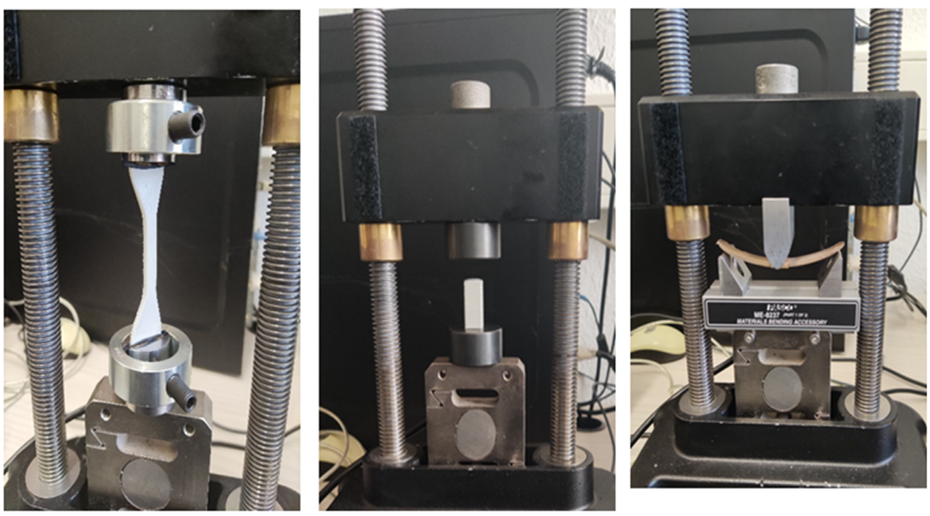
The BPp reinforced rPP blend composites and PP were subjected to tensile tests using a PASCO ME-8236 material test machine according to ASTM D638 Type IV, and the samples were designed and printed according to the same standard. The tensile properties of the material were measured at room temperature with the test speed set at 5 mm/min to obtain the strength. According to **Figure 7(a)**, the ends of the test specimen are fixed in the clamps of the test device. After that, the load increases steadily. Three samples were tested, and the average value of the tests was taken as the result.

2.4.2. Compression Test

The compressive properties of BPp/rPP composites were measured according to ASTM D695, the compression test was conducted at a speed of 2 mm/min, and the compressive strength was evaluated using the same universal testing machine as the tensile test. The samples were placed between compression plates parallel to the surfaces shown in **Figure 7(b)**. The sample was then uniformly compressed. Average value of three samples evaluated for compressive strength of PP and BPp reinforced rPP composites. The compressive strength was determined at a deformation of 10% of the original height of the specimen.

2.4.3. Flexural Test

The ability of a biocomposite material to withstand bending forces applied perpendicular to its longitudinal axis is called flexural strength. The flexural property of the BPp/rPP composite and PP was measured according to ASTM D790, which has a three-point bending configuration **Figure 7(c)**. The bending test was conducted at a speed of 2 mm/min to evaluate the bending strength, this was accomplished using the same machine that conducted the tensile test. Compressive



(a) (b) (c)

Figure 7. (a) Layout of the tensile test specimen; (b) Layout of the compression test specimen; (c) Layout of the flexural test specimen.

strength was determined when the maximum strain of 5% was reached. The average values of three samples were calculated.

Calculate the flexural strength using the following formula:

$$\sigma = \frac{3FL}{2wt^2} \tag{1}$$

where:

F—Max load (N),

L—Distance between the supports (mm),

w—Width of the Specimen (mm),

t—Thickness of the specimen (mm).

The bending stress was determined as follows:

$$\varepsilon = \frac{6\mathrm{d}t}{L^2} \tag{2}$$

where, d is the deflection, *t* is the thickness of the specimen (mm) and *L* is the distance between the supports (mm).

4. Results and Discussions

4.1. Surface Roughness Analysis

Average values of the surface roughness (Ra, Rz) of the filaments are presented numerically in **Table 4** and graphically in **Figure 8**.

As we can see, the surface roughness of the rPP filament is significantly lower than that of the other two BPp reinforced filaments. The quantity of reinforcement affects the surface roughness. Considering the Ra and Rz values of the roughness of recycled polypropylene filaments, which are 0.5720 µm and 27.14 µm, respectively, to understand the dependence of the roughness on the fiber amount. On the other hand, Ra and Rz steadily rise in filaments reinforced with BPp proportions; for the 5-rPP filament, they reach values of 2.287 µm and 31.38 µm, respectively, and for the 10-rPP filament, they reach values of 2.397 µm and 33.90 µm, respectively. The hydrophilic character of bean pod powder and the hydrophobic nature of rPP contribute to the increase in roughness of BPp/rPP filaments, also the size of the BPp particles explains the increase in roughness.

4.2. Particles Size Analyses

The particle size distributions of the BPp were presented in **Figure 9**. When the particle size distribution of the BPp was observed, the $Q_3(10)$, $Q_3(50)$ and

Table 4. Average surface roughness values of the filaments.

Filaments	Ra (µm)	Rz (µm)
rPP	0.5720	27.14
5-rPP	2.287	31.38
10-rPP	2.397	33.90

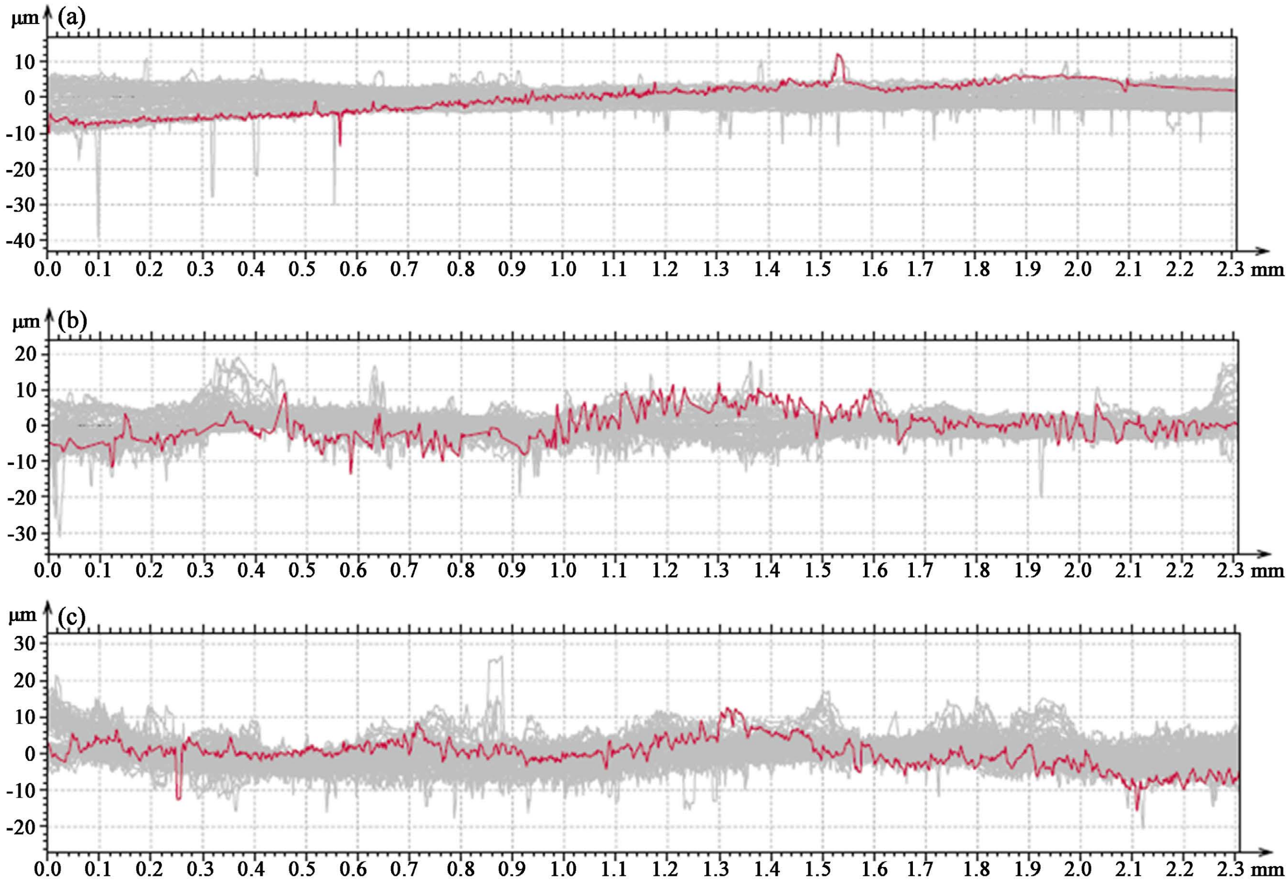


Figure 8. (a) surface roughness of rPP, (b) surface roughness of 5-rPP, (c) surface roughness of 10-rPP.

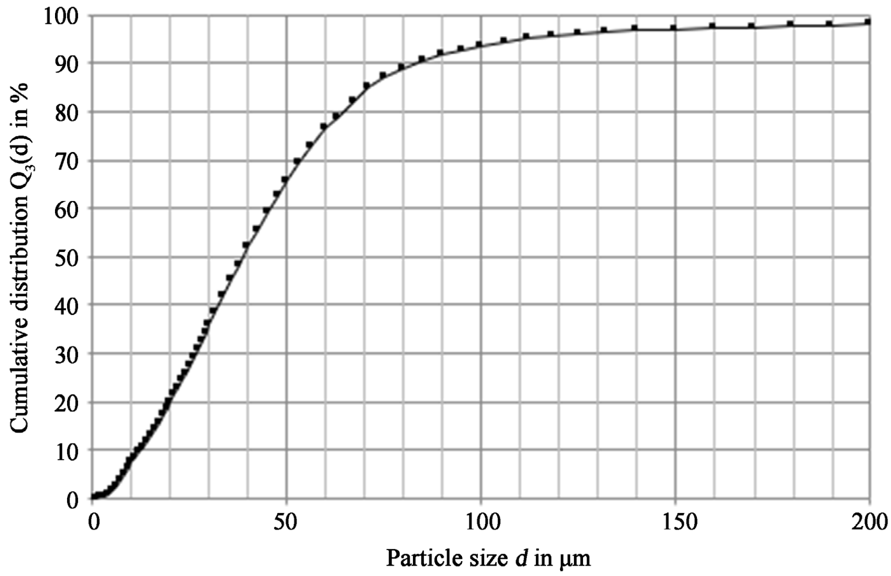


Figure 9. Particle size analysis.

$Q_3(90)$ values of the bean pod powder were determined to be 12.8, 39.9 and 83.28 µm, respectively. Particle size distribution could also explain the smooth and rough surface of composite filaments.

4.3. Tensile Strength

Figure 10 shows the tensile strength of pure PP, rPP and the effect of BPp loading on the tensile strength of rPP. Table 5 shows the tensile strength value of pure PP, rPP and BPp-PP composites.

Biocomposites' tensile properties are generally influenced by the filler content, and as the filler amount increases, so does the mechanical performance. The tensile strength of the neat PP and rPP test samples, printed using the FFF process, was 18.1 MPa and 20.4 MPa, respectively. According to the curves, less BPp can increase the tensile strength of the composites, as the amount of BPp increases further, the tensile strength of the composites starts to decrease. The BPp/rPP composite with a BBp content of 5 wt% has the best tensile strength of 22.8 MPa among the BPp/rPP composites. The excellent BPp dispersion in the rPP matrix may be the cause. In fact, the rPP could only completely enclose a smaller amount of the powder from the bean pods, which drew the two together. However, the tensile strength started to decline beyond the BPp weight fraction of 5 wt% in the rPP. The increased surface area interaction of the matrix molecules with the filler particles can be used to explain the decrease in tensile strength. The polar fillers and the non-polar matrix have a large interfacial area because the fillers are so tiny. The weakening of the link between the two causes the tensile strength to diminish as this interfacial area rises [36]. With increasing

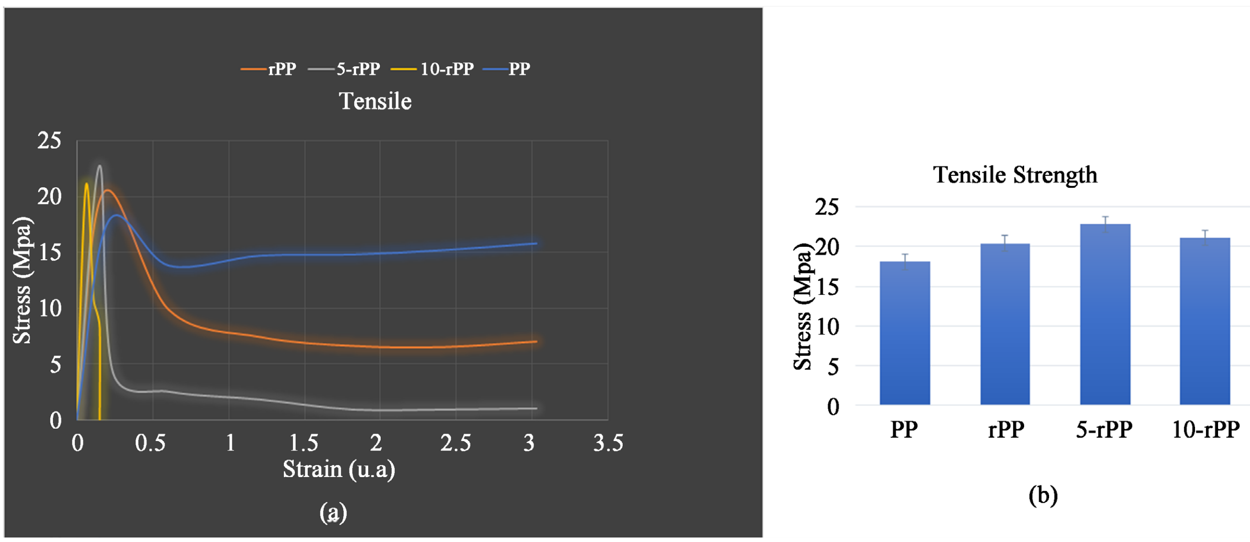


Figure 10. (a) Tensile test; (b) Tensile strength.

Table 5. Mechanical properties of the neat PP, rPP, BPp/rPP composites.

	TS (MPa)	FS (MPa)	CS (MPa)
PP	18.1	9.78	20.5
rPP	20.4	11.09	22.6
5-rPP	22.8	15.05	24.2
10-rPP	21.1	14.2	24.5

fillers, areas of stress concentration in the matrix can occur due to the sharp angles of irregularly shaped filler particles during tensile loading [37].

4.4. Flexural Strength

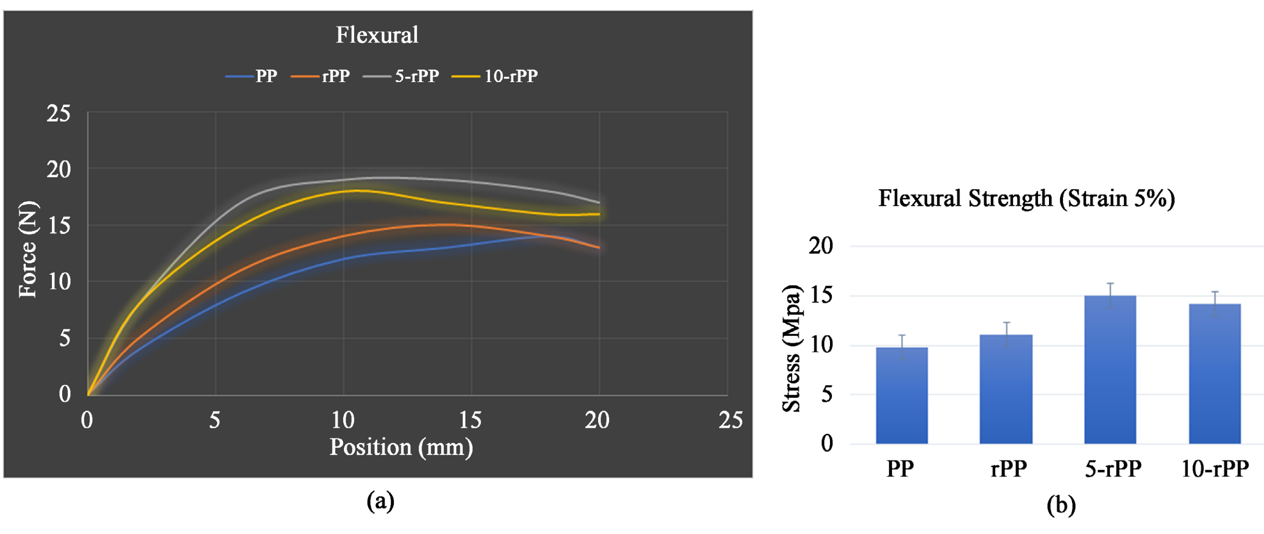
For a better understanding of the mechanical properties of BPp/rPP composites, a Flexural test was also performed. The flexural properties of pure PP, rPP and BPp/rPP composites at different loads are presented in **Figure 11**. The flexural strength values of pure PP, rPP and its biocomposites with BPp are shown in **Table 5**.

It was found that pure PP and rPP have a flexural strength of about 9.78 MPa and 11.09 MPa, respectively. The flexural strength values of rPP composites containing bean pod powder were higher than those of pure PP and rPP. The maximum flexural strength is achieved with the composite prepared with 5% BPp, which is about 15.05 MPa. The presence of toughness enhancing substances such as: silica, iron and others in the BPp increases the stiffness of the composites. Above 5 wt% BPp, the flexural strength result shows that increasing the volume of BPp in the matrix rPP make decreases the flexural strength of the composite material, it follows that the volume fraction of BPp tends to affect the strength of the biocomposite, which may be related to agglomeration. The flexural strength also depends on the composition of the fiber.

4.5. Compression Strength

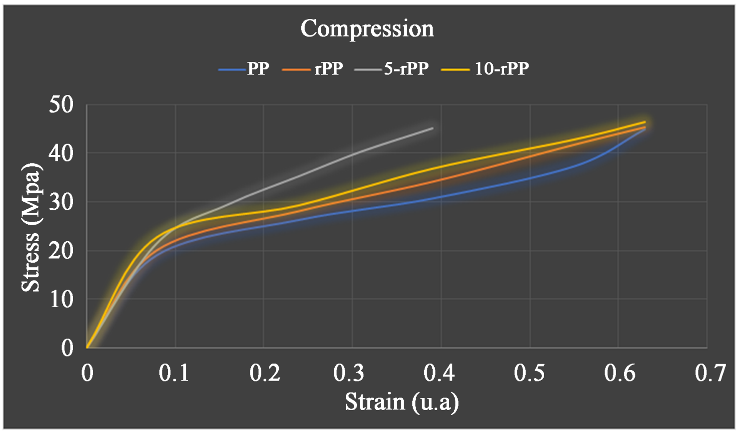
Figure 12 shows the compressive strength of pure PP, rPP and BPp/rPP composites under different BPp loadings. The compressive strength values at 10% strain of pure PP, rPP and its BPp reinforced composites are also presented in **Table 5**.

The compressive strength of pure PP and rPP was 20.5 and 22.6 MPa respectively. After mixing 5 and 10 wt% BPp, the compressive strength value increased to 24.2 and 24.6 MPa. From these values, it can be noted that the compressive

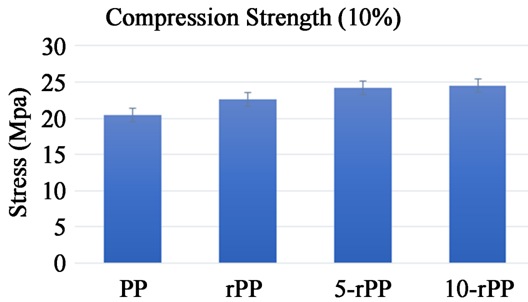


(a) (b)

Figure 11. (a) Flexural test; (b) Flexural strength.



(a)



(b)

Figure 12. (a) Compression test; (b) Compression strength.

strength increases as the BPp load increases. The compressive strength of the BPp/rPP composite is high because the bean pods powder fills the space in the chains and pores of the rPP. In addition, the compressive strength is high also because of the effective stress transfer, meaning that there is a strong bond between rPP and BPp.

5. Conclusion

This study demonstrated the feasibility of using BPp as agricultural waste and rPP as plastic waste in the manufacture of composites. It was observed that BPp in rPP improves the mechanical properties of the biocomposite. The tensile strength, flexural strength, and compression strength of rPP composites containing BPp were higher than those of rPP and pure PP. Among the BPp/rPP composites, the best tensile strength and flexural strength were for 5-rPP which are 22.8 MPa and 15.05 MPa respectively. The best compression strength was that of 10-rPP which were 24.5 MPa. Above the BPp weight fraction of 5 wt% in rPP, the tensile and Flexural strength began to decrease, and the surface of the filaments became increasingly rough as the amount of BPp increased. This study showed that bean pods powder can be used with rPP as a reinforcing filler. The bean pods particles have a great potential to improve the mechanical properties of rPP. However, other investigation can be made in the use of bean pods in the manufacture of composites such as: chemical treatment of the bean pods, use of coupling agents and elastomer impact modifiers to improve compatibility and optimization of extrusion and printing parameters.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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