Investigation Influence of Styrene Butadiene Rubber on Physical, Mechanical and Thermal Properties of Polypropylene Composite for 3D Printing Filaments

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ABSTRACT

The increasing amount of tire waste has become a global environmental problem due to its non-biodegradable nature and potential to pollute the air, soil, and water. One solution is to recycle the tire waste into Styrene Butadiene Rubber (SBR) as a filler in polymer composites for 3D printing filaments. This study aims to analyze the effect of adding SBR on the physical, mechanical, and thermal properties of Polypropylene (PP) composites. The methods used included extrusion and filament molding with variations in SBR mass fraction: 0%, 5%, 10%, and 15%. Density testing, tensile testing, thermal analysis, and morphological observation were conducted. The results show that theoretically, the density of the composite increases with the addition of SBR, but in practice, the density decreases due to the formation of voids. The increase in porosity also affects the mechanical properties. The tensile tests show an 8.8% increase in the tensile strength with a 5% addition of SBR, but a significant decrease with 10% and 15% additions due to the particle agglomeration and poor interfacial adhesion. The thermal stability decreases slightly with the addition of SBR, as seen from the earlier degradation and increased post-Thermogravimetry Analysis (TGA) residue. The surface morphology becomes rougher with an increase in the SBR fraction, affecting the quality of the 3D prints. It can be concluded that the addition of up to 5% SBR improves the mechanical properties, whereas further increases in the fraction result in a decline in

the material performance. This study demonstrates the potential for utilizing tire waste in the production of PP/SBR composites for 3D printing applications with the necessary formulation and process optimization.

Keywords-Styrene Butadiene Rubber; Polypropylene; tensile strength; thermal stability; 3D printing; density; porosity; surface morphology

I. INTRODUCTION

The increasing amount of tire waste generated into the environment each year is a global problem [1]. The annual global production is estimated to reach 1.4 billion units, equivalent to 17 million tons of used tires [2]. Most developed countries manage the municipal solid waste effectively, but the low-income and lower-middle-income countries face significant challenges due to the inadequate waste management infrastructure, informal sectors, and a lack of trained personnel [3]. Tire waste is known as "black pollution" due to its low recycling rate and potential to cause environmental problems if not handled properly [4]. This is exacerbated by the fact that tires are highly flammable. Uncontrolled open tire fires produce a number of air pollutants, such as particulates, carbon monoxide (CO), sulfur oxides (SO_2), nitrogen oxides (NO_x), and Volatile Organic Compounds (VOCs). Emissions from burning the used tires can pose chronic and long-term health hazards to people living near the burning site [5]. At landfills, the tire waste requires a big space due to its shape and size. In addition, the used tires can accumulate in water, providing an ideal breeding ground for disease-carrying pests, such as mosquitoes [6].

Used tires can be recycled into valuable raw materials, such as Ground Tire Rubber (GTR), for use in the construction industry and partial replacement in pure rubber compounds, offering a sustainable solution for the environment and circular economy [7, 8]. Research has investigated the use of GTR products as fillers for cement mortar, other construction materials, asphalt, and polymers [9]. Numerous studies have examined the incorporation of GTR into thermoplastics, thermosets, and rubber. GTR has been added to thermoplastics, such as PP, Low-Density Polyethylene (LDPE), High-Density Polyethylene (HDPE), Polyvinyl Chloride (PVC), and others. Typically, the addition of GTR to polymers leads to the degradation of their mechanical properties [7].

The need for environmental protection and green energy has driven the development of polymer composites that use recycled materials [10]. Polymer composites are a combination of two or more polymers with different characteristics. At certain ratios, this combination allows for a synergy between various properties, such as physicochemical, thermal, and separation capabilities in the resulting material [11].

3D Printing composites have been used in a variety of applications, including biomedical, mechanical, electrical, thermal, and optically enhanced products. The growing popularity of 3D Printing composites can be attributed to their ability to create complex geometries, low-cost production, and other advantages associated with rapid prototyping [12]. PP has a good flexural strength and moisture resistance, especially when used in composites with wood fibers or other fibers [13]. PP 3D printing filaments can undergo shrinkage and warping, as well as process-related morphological and crystallographic changes [14]. The objective of this study is to analyze PP composites with SBR fillers to produce 3D printing filaments that can be used to the manufacture of functional products and prototypes. In this study, SBR from recycled tire waste was used as the filler, and PP was utilized as the matrix. The mechanical strength of the composite was determined through tensile testing, its physical properties through density testing and Scanning Electron Microscopy (SEM), while its thermal properties through TGA testing.

II. MATERIALS AND METHODS

A. Materials

The PP used was LyondellBasell Moplen HP500N, with a density of 0.9 g/cm^3, and the SBR had a mesh size of 40. Extrusion was performed with an extrusion rate of 10"/min-26"/min, nozzle diameter of 1.75 mm, and a maximum working temperature of 160°C. The mass fractions of SBR were 0%, 5%, 10%, and 15%, with a mesh size of 80/100. The properties of PP and SBR are presented in Table I.

TABLE I. PROPERTIES OF PP AND SBR

Properties	PP	SBR
Melt flow rate	1 g/min	
Density	0.9 g/cm^3	
Tensile strength	34 MPa	
Melting temperature	163°C	
Particle size	Mesh 40	Mesh 40
Hardness		59 Shore A
Carbon black		28%
Acetone extract		14%

B. Methods

Specimens were produced using a Creality K1 3D printer with a print volume of 220 mm × 220 mm × 250 mm. The specimen printing parameters are described in Table II, considering the test results and the parameters from [15]. The infill pattern used a grid pattern with the addition of a brim, as portrayed in Figure 1.

TABLE II. PARAMETER PRINTING

Parameter	Value
Nozzle diameter	1 mm
Layer thickness	0.32 mm
Infill degree	100%
Printing speed	20 mm/s
Bed temperature	100°C
Nozzle temperature	270°C
Infill pattern	Grid

C. Experimental Methods

Density testing was conducted to obtain the density of the PP/SBR composite, in accordance with ASTM D792. Theoretical density calculations were performed in accordance with the rule of mixture, while actual calculations were performed experimentally in a 96% solvent/ethanol solution. The tensile test specimens, as depicted in Figure 2, were molded in accordance with ASTM D638 Type 5 standard. The molded specimens were tested using a JTM-UTS510 tensile testing machine at a loading rate of 10 mm/min.

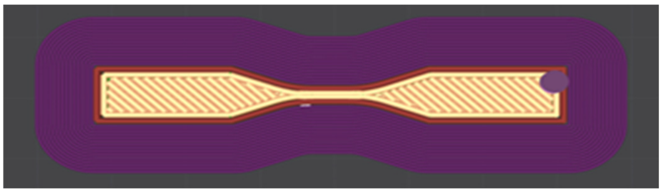


Fig. 1. Tensile test specimen printing scheme for 3D printing.

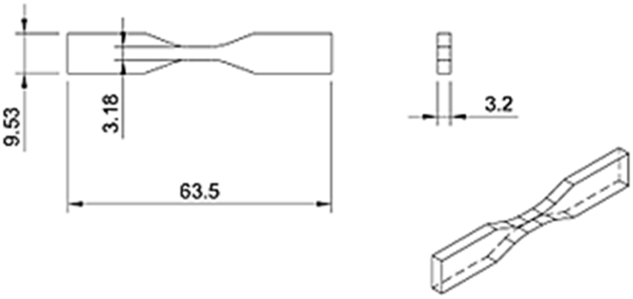


Fig. 2. Tensile test specimen dimensions (mm).

The TGA test was conducted by taking several samples from different areas and either combining them for a single determination or running each separately, with the final analysis representing the average determination. Samples were taken with a weight of 5 mg-10 mg, a flow rate of 10 °C/min, a purge time of 2 min, and a nitrogen atmosphere. The SEM data acquisition used fracture samples from tensile testing to examine their morphological structure (cross-section). The PP composite samples were mounted on a sample holder using double-sided adhesive tape and then coated with platinum in a vacuum. Topographic images were captured at 200× magnification with an acceleration voltage of 15 kV.

III. RESULTS AND DISCUSSION

A. *Density and Porosity*

Density testing was conducted to determine the effect of different SBR contents on the density of the composite. The results of density testing of the PP composites containing SBR are shown in Figure 3. In theory, the addition of SBR should increase the density of the composite. The density of a composite is influenced by the matrix and filler used. If the density of the fiber is lower than that of the matrix, then, theoretically, the composite will have a lower density, and vice versa [16, 17]. The density test results show values close to the theoretical calculations. However, differences may arise due to external factors, which can affect the formation of voids in the composite, ultimately causing differences between the theoretical and actual densities. Figure 3(a) shows that the theoretical density value of PP/SBR is higher than the experimental or actual density value.

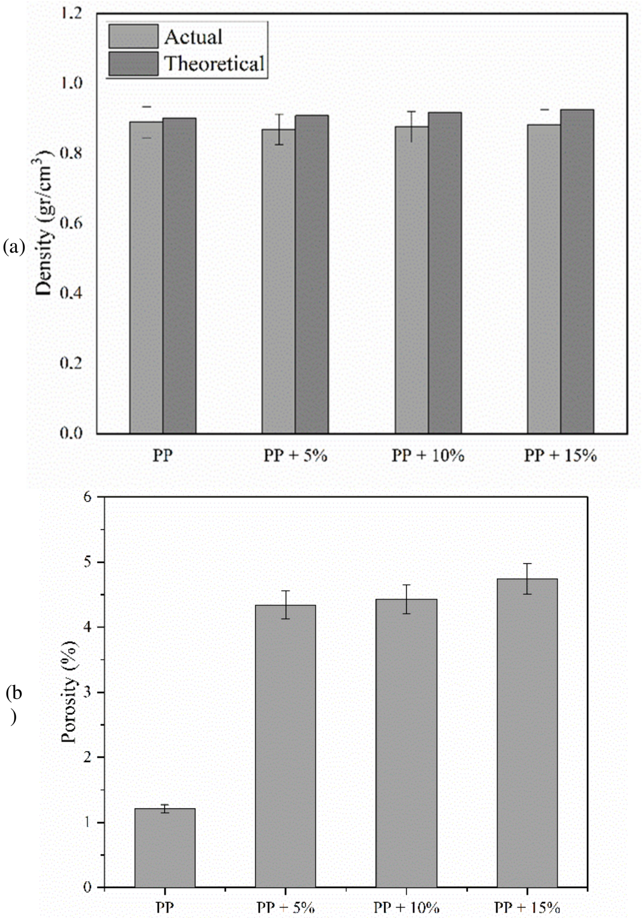


Fig. 3. The effect of SBR addition on: (a) density, (b) porosity of PP.

The density of the composite increases with the addition of SBR, but this increase is also accompanied by an increase in voids. This decrease in density is caused by the presence of air voids [9]. According to the Macromorphology test results, the voids tend to be more easily identified because they affect the mechanical properties of the composite. The calculated porosity percentages for each variation were: PP (1.21%), PP + 5% SBR (4.34%), PP + 10% SBR (4.43%), and PP + 15% SBR (4.74%), as presented in Figure 3(b). Another contributing factor is the suboptimal quality of the filaments, as indicated by the instability of the diameter of the filaments produced using the extruder machine. In addition, the density of the specimen is influenced by the production process using 3D printing. Imperfections in printing parameters, such as suboptimal extrusion temperature or uneven filament flow rate, can cause gaps in the mold structure, which contribute to an increase in the number of voids [18].

B. *Tensile Strength*

PP-SBR specimens were tested for tensile strength in accordance with ASTM D638 Type 5 standards. The tensile test results are illustrated in Figure 4. Based on the graph, the tensile strength value for pure PP was 27.42 MPa. This value increased to 29.82 MPa for PP + 5%, but decreased for PP + 10% (25.42 MPa) and PP + 15% (23.08 MPa). Meanwhile, the Young's modulus for pure PP was 0.27 GPa, then increased to 0.45 GPa for PP + 5%, but decreased to 0.35 GPa for PP + 10% and 0.24 GPa for PP + 15%. These results are consistent with [19], where it is shown that adding less than 5% SBR results in the highest tensile strength.

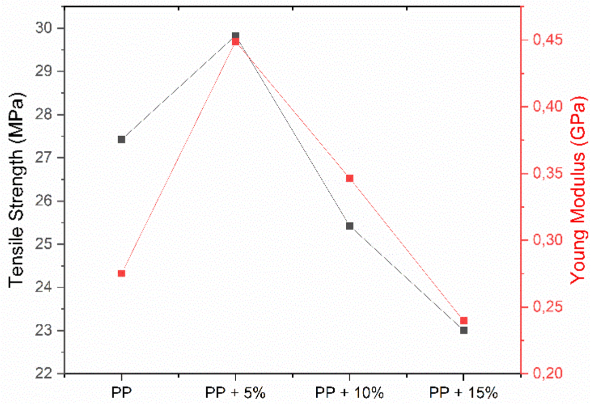


Fig. 4. Tensile strength of PP/SBR.

The results illustrated in Figure 4 indicate that the addition of 5% SBR increased the tensile strength by 8.8% compared to the pure PP. However, there is a decrease of 7.3% in the 10% SBR fraction and a more significant decrease of 16.1% in the 15% SBR fraction compared to the pure PP. The increase in the tensile strength in PP + 5% is due to the uniform distribution of the irregularly shaped rubber particles with rough surfaces and numerous voids, which contribute to better adhesion [20], although they also cause the formation of voids. However, at 10% and 15% variations, the tensile strength decreased significantly. This decrease was caused by the formation of voids in the material and reduced the interfacial adhesion between the rubber particles and polymers. These results are consistent with the findings reported in [21, 22], which revealed the appearance of voids in PP due to the influence of SBR. A higher SBR content can also cause particle agglomeration, which contributes to weak interfacial bonds between the filler and matrix, thereby reducing the tensile strength [9].

Young's modulus is a mechanical property of a material that indicates its stiffness and resistance to elastic deformation, as well as its ability to return to its original size after being subjected to stress [23]. The addition of SBR up to 5% increased the Young's modulus, indicating an increase in the material stiffness. This is due to the rubber particles acting as fillers, thus inhibiting the deformation of PP. However, at SBR fractions of 10% and 15%, the Young's modulus decreased. This decrease is caused by the dominance of the softer elastomeric properties of rubber, making the material more flexible and less rigid [9].

C. *Thermogravimetric Analysis*

TGA testing is used to evaluate the material stability based on the mass loss due to the decomposition or combustion processes resulting from the addition of SBR to composites [24]. Figure 5(a) shows the TGA test result graph for the composite. The TGA graph exhibits the thermal degradation pattern of the PP composites with varying mass fractions of SBR. From the results obtained, it can be observed that all samples have similar thermal degradation patterns, with significant weight loss occurring in the temperature range of approximately 400–500°C. This indicates that the presence of SBR in the composite does not drastically alter the thermal degradation mechanism of PP, but only slightly lowers the onset temperature of degradation. Crumb rubber-based materials in Acrylonitrile-Butadiene-Styrene copolymer (ABS) composites also exhibit slightly earlier thermal degradation compared to the pure polymers [24]. Figure 5(b) shows the Differential Thermogravimetry (DTG) curve from the TGA testing of the composite.

The main thermal decomposition in the graph occurs at a temperature of around 450°C, which shows the characteristic of PP undergoing polymer chain scission due to high temperature. The addition of rubber particles can contribute to slight changes in the thermal stability of the composite [24]. In this study, with the increase in the mass fraction of SBR, there was a slight shift in the degradation curve to a lower temperature. The TGA thermogram of the rubber particles demonstrated that the material had a lower thermal stability triggering earlier degradation. The thermal changes in PP/SBR materials are outlined in Table III.

TABLE III. TGA AND DTG OBSERVATION RESULTS

SBR fraction	**Onset**	**Peak**	**End-set**
0%	390 °C	445 °C	482 °C
5%	388 °C	450 °C	485 °C
10%	385 °C	455 °C	488 °C
15%	380 °C	460 °C	490 °C

In addition, the amount of the residue remaining after heating to around 600°C also increased with the addition of SBR in the composite. This residue is associated with the mineral content or inorganic fillers derived from the used tire fraction [24]. The increase in the amount of residue in this study indicates that SBR does leave behind material that is not easily vaporized during the thermal degradation process.

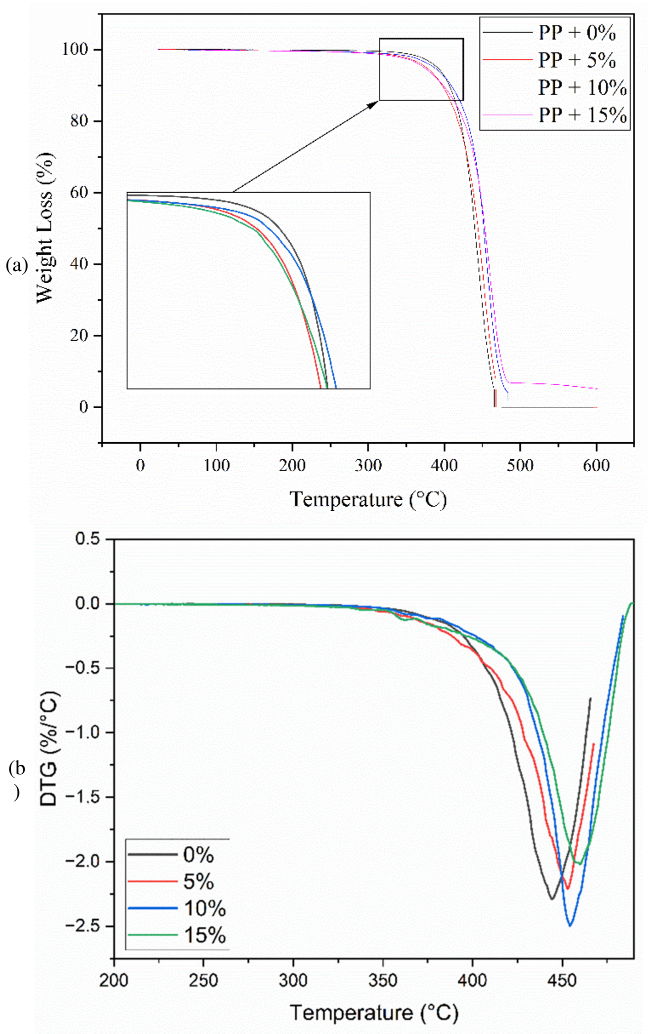


Fig. 5. The effect of SBR on: (a) thermogravimetric analysis, (b) DTG of PP.

D. *Macromorphology and Scanning Electron Microscope*

Based on the results of the macromorphology, the variations in the mass fraction of SBR affect the dimensions and morphology of the printed filaments. The results of the morphology test of the PP composite containing SBR observed under the macro microscope are shown in Figure 6.

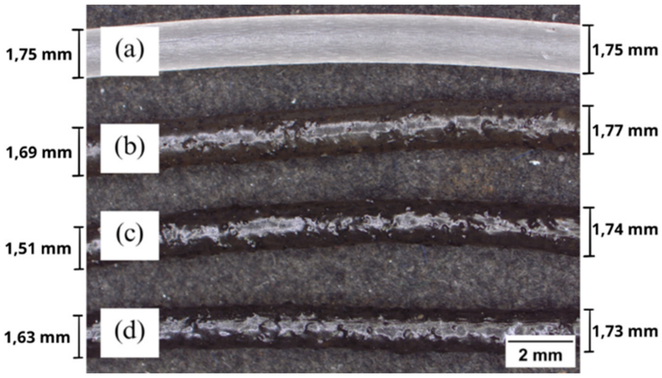
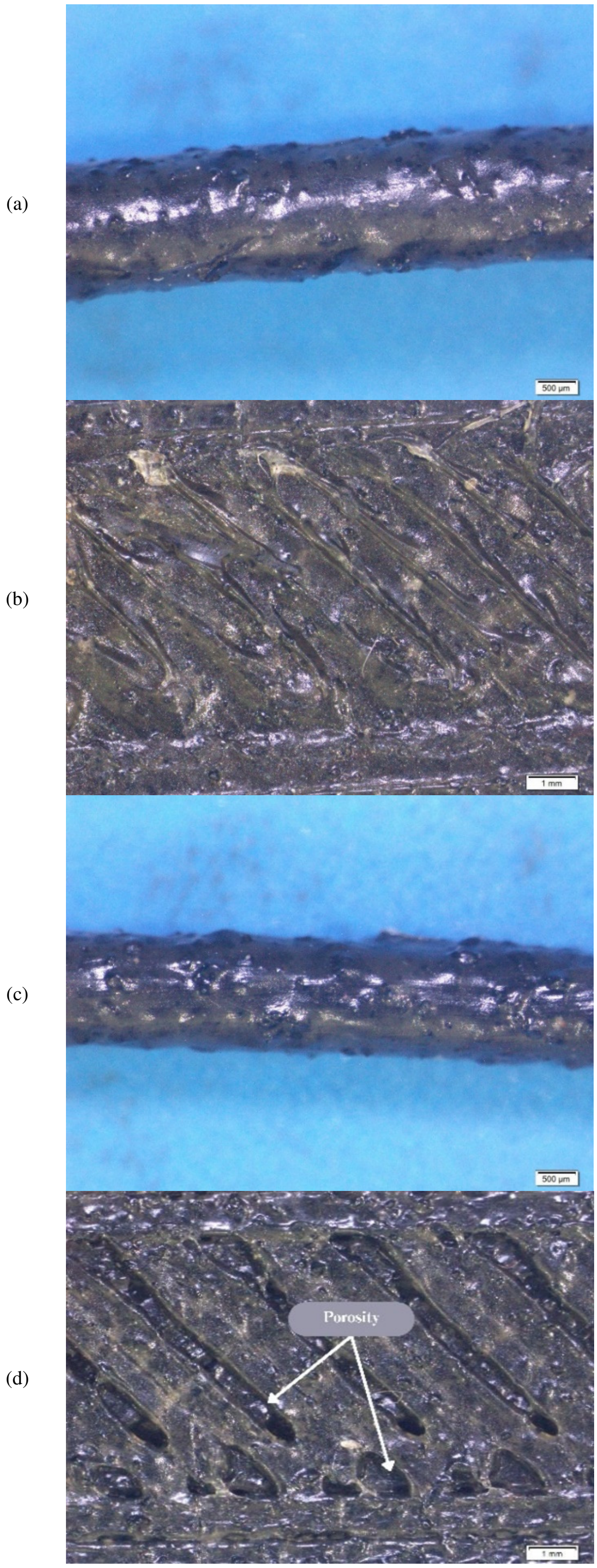


Fig. 6. Results of macrofilament microscope observations with added fractions of SBR: (a) PP, (b) PP + 5%, (c) PP + 10%, (d) PP + 15%.



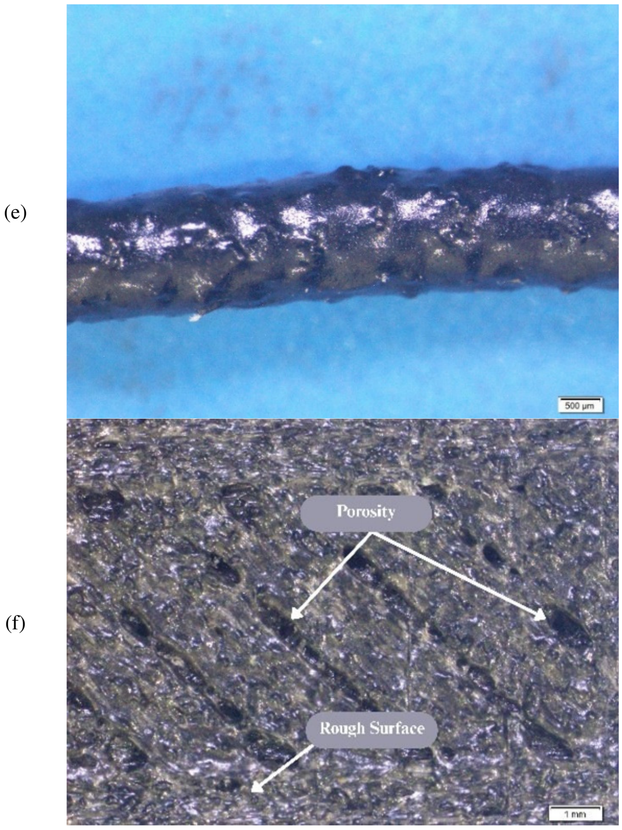


Fig. 7. Macro filament observation and 3D printing results: (a-b) 5% SBR, (c-d) 10% SBR, (e-f) 15% SBR.

The addition of SBR to the PP/SBR composite filaments caused significant changes in the morphology and dimensions of the filaments and 3D printed specimens. As the rubber content increases, the surface roughness and morphology of the composite change [15]. This is in line with [15], where it was stated that the addition of fillers to the PP results in increased the surface roughness of the filaments. In the filaments, increasing the SBR mass fraction from 5% to 15% resulted in increasingly rough surfaces and less uniform diameters, as shown in Figure 7(a), (c), and (e). This surface roughness and diameter variation can affect the quality of the 3D prints [25]. The specimens with 5% SBR showed a fairly regular layer structure, although there were some imperfections in the printing process, as depicted in Figure 7(b). However, at SBR mass fractions of 10% and 15%, the specimen surface became rougher with an increase in the number of pores and irregularities in the distribution of material, as illustrated in Figures 7(d) and (f). The presence of voids affects the overall mechanical properties of the specimens produced [26].

IV. CONCLUSION

The addition of Styrene Butadiene Rubber (SBR) to Polypropylene (PP) composites significantly alters the physical, mechanical, and thermal properties of the material. Although the theoretical density increases with the SBR content, the experimental measurements show reductions due to the void formation, complying with prior studies on polymer-rubber blends, while 5% SBR increases the ultimate tensile strength and Young's modulus. However, higher concentrations (10% and 15%) degrade these properties due to the rubber particle agglomeration and poor interfacial adhesion, a phenomenon also observed in incompatible polymer systems. The thermal stability slightly decreases, with the main degradation occurring around 450°C, aligning with earlier studies demonstrating the rubber's impact on the thermal performance. Additionally, the changes in the surface morphology, including increased roughness and uneven filament diameter, negatively affect the 3D print quality.

A unique contribution of this study is the detailed characterization of SBR's influence on porosity, interfacial adhesion, and printability. Nevertheless, limitations, such as poor interfacial bonding, highlight the need for compatibilizers like maleic anhydride-grafted PP (MA-g-PP), alternative rubbers to improve the thermal stability, and further exploration of the processing parameters for property optimization. Future research should also incorporate Dynamic Mechanical Analysis (DMA) and long-term durability studies to enhance the understanding of the viscoelastic behavior and real-world applicability.

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DATA AVAILABILITY

Dataset available on request form the corresponding author

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