

Research Paper

## Analysis of the Effect of Adding *Jatropha* Seed Shell on the Physical and Thermal Properties of Recycled Polypropylene Composites

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**ABSTRACT:** The increasing volume of plastic waste, particularly polypropylene (PP), poses a significant environmental challenge. Recycling is a crucial solution but often leads to degradation of PP's mechanical and thermal properties of PP. This study aims to enhance recycled PP quality by adding a natural filler in the form of *Jatropha* Seed Shell (JSS). JSS is a potential filler due to its good mechanical and thermal properties, low density, and recyclability. The research was conducted in two parts: first, without alkalization treatment, using recycled PP with varying JSS additions at 0%, 5%, 10%, 15%, and 20%. The Melt Flow Rate (MFR) test without JSS showed an average value of 20.620 g/10 minutes, decreasing to 15.380 g/10 minutes at 20% JSS. JSS addition also increased crystallinity from 30.92% to 36.14% and melting temperature from 163.9°C to 166.4°C with 20% JSS. The second part, with alkalization treatment using a 20% NaOH solution, aimed to improve JSS compatibility with the PP matrix. The results showed that alkalization treatment improved the distribution of the filler within the matrix, leading to enhanced mechanical and thermal properties. The MFR value decreased more significantly in this treatment compared to without alkalization, with the lowest value at 14.55 g/10 minutes in the composite with 20% JSS. The degree of crystallinity increased to 33.9% with 15% JSS addition, while the melting temperature remained stable.

**Keywords:** Alkalization, Degree of Crystallinity, *Jatropha* Seed Shell, MFR, Recycled Polypropylene

## 1. INTRODUCTION

Environmental issues caused by plastic waste remain a global concern, primarily due to the non-biodegradable nature of plastics. Polypropylene (PP) is one of the most widely used plastics, known for its advantages such as good mechanical strength, chemical resistance, and relatively low production costs. However, the high consumption of PP in various applications has led to a significant increase in PP waste volume, posing major challenges in its management [1]. Recycling plastics is an effective solution to mitigate the negative impacts of plastic waste on the environment. However, recycling PP often results in a decline in material quality, such as degradation of mechanical and thermal properties, due to repeated heating and polymer chain scission. To address this degradation, one viable approach is to modify recycled PP material using environmentally friendly natural fillers [1] [2].

*Jatropha* Seed Shell (JSS) is a natural material with potential as a filler in polymer composites [3]. *Jatropha* Seed Shell (JSS) contains components such as cellulose, hemicellulose, and lignin, which can enhance the mechanical and thermal properties of composite materials. Additionally, JSS has a low density, allowing for weight reduction in composite materials without compromising strength. Several previous studies have explored the use of Castor Bean Shell as a filler in polymer composites to improve the mechanical and thermal properties of materials. Ogah conducted tensile strength tests on polyester/Castor Bean Shell composites. The highest tensile strength value was achieved with a 15% addition of Castor Bean Shell, measuring 50 MPa [4].

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Dara's research showed that tensile strength and hardness testing on an aluminum matrix with a 12% addition of Castor Bean Shell significantly increased compared to pure aluminum, with tensile strength improving by approximately 24% from 124.23 MPa to 154.03 MPa and hardness increasing by 43% from 95.9 MPa to 137.3 MPa [3]. Tensile strength is related to the melt flow index (MFI). A low MFI can result in high tensile strength. Nurhajati [5] reported the decrease in MFI from 7 g/10 minutes to 2.5 g/10 minutes impacted tensile strength, where the tensile strength increased from 51.96 MPa to 63.25 MPa. A lower MFI value contributes to higher tensile strength. The JSS as a filler not only has the potential to improve the quality of recycled polymers but also supports the utilization of more sustainable and environmentally friendly materials [6].

This study offers a novel contribution by investigating the utilization of Jatropha Seed Shell (JSS) as a filler in recycled polypropylene (PP)-based composites, an area that has not been extensively explored in previous research. Unlike prior studies that primarily focused on Castor Bean Shell (CBS) or other natural fillers, this research highlights the unique properties of JSS, including its cellulose, hemicellulose, and lignin content, and its potential to enhance the mechanical and thermal properties of recycled PP composites. The study introduces two key novelties: the integration of alkalization treatment to improve compatibility between JSS and the recycled PP matrix and the optimization of JSS concentration for optimal composite performance.

The application of alkalization treatment is a novel approach, expected to improve filler dispersion and interfacial bonding within the composite material. This treatment aims to enhance the overall properties of the composite by ensuring better interaction between the filler and the polymer matrix. Additionally, the research focuses on systematically determining the optimal concentration of JSS filler in recycled PP-based composites, balancing enhancements in mechanical and thermal properties with the material's sustainability and lightweight characteristics. By utilizing recycled materials and renewable natural resources, this study is expected to contribute to the advancement of sustainable composite technologies, addressing the dual challenges of plastic waste reduction and the development of eco-friendly materials for industrial applications.

## 2. RESEARCH METODOLOGY

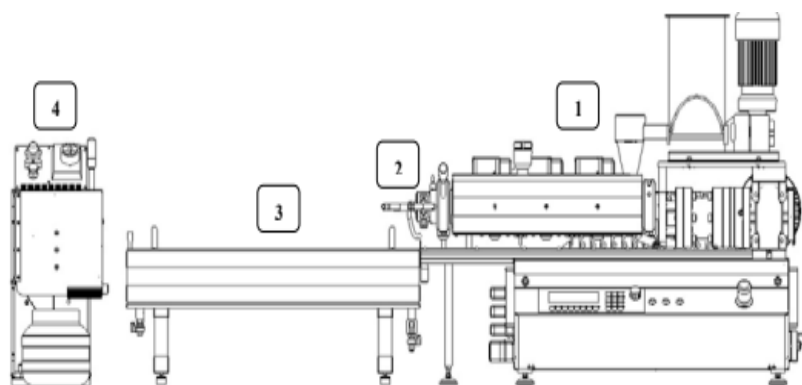
### 2.1. Tools and Materials

The equipment used in this study includes an oven, blender, 80-mesh sieve, analytical balance, Melt Flow Index Dynisco 5000 series, Differential Scanning Calorimetry 214 Polyma, Compounder Teach-Line ZK 25 × 24 D, aluminum crucible, beaker glass, and sieve shaker. The materials used are recycled polypropylene from Surya Indo Utama, Jatropha seed shell, analytical grade NaOH, and distilled water.

### 2.2. Research Methods

This study aims to develop a composite based on recycled polypropylene (rPP) by incorporating Jatropha Seed Shell (JSS) as a filler to enhance the material's mechanical and thermal properties [7]. The research process begins with material preparation, where JSS is sun-dried for two days, followed by oven-drying at 110°C until a constant weight is achieved to ensure the material is free from moisture. A portion of the JSS is then immersed in a 20% NaOH solution for one hour as part of an alkalization treatment, which is intended to improve the compatibility between the filler and the rPP matrix [2]. After soaking, the JSS is rinsed with water until neutral pH is achieved, oven-dried again, ground into fine powder using a blender, and sieved through an 80-mesh sieve shaker [8].

The composite manufacturing process is carried out using a Twin-Screw Extruder, with the barrel zone temperature set at 190°C and a screw rotation speed of 35 rpm to ensure homogeneous mixing of rPP and Jatropha Seed Shell (JSS) [9]. The mixture is prepared with varying Jatropha Seed Shell (JSS) compositions of 0%, 5%, 10%, 15%, and 20% by weight, with a total sample weight of 200 grams. Once the mixing process is complete, the composite melt is cooled in a water bath and cut into pellets using a pelletizer. The main tool design is shown in Figure 1.



**Figure 1.** Twin Screw Extruder.

1. Ekstruder, 2. Strand die, 3. Water bath, 4. Strand pelletizer

### 2.3. Analysis Methods

Characterization tests on the composite are Melt Flow Rate (MFR) testing using a Melt Flow Indexer (MFI) according to ASTM D1238 standards) and a Differential Scanning Calorimeter (DSC) following ASTM D3418 standards to measure melting temperature ( $T_m$ ) and degree of crystallinity ( $X_c$ ). Melt Flow Rate (MFR) testing is a method used to measure the flowability of thermoplastic materials when melted under specific conditions. It provides insight into the viscosity and processability of the material. The test is conducted using a Melt Flow Indexer, where a specific amount of polymer is melted and extruded through a capillary die under a standard weight and temperature. Differential Scanning Calorimetry (DSC) is a thermal analysis technique used to measure the heat flow associated with material transitions, such as melting, crystallization, and glass transition. The test provides data on the material's thermal properties, such as melting temperature ( $T_m$ ), crystallization temperature ( $T_c$ ), and degree of crystallinity [10] [11].

The results from the MFI and DSC tests are analyzed to evaluate the effect of adding JSS, both with and without alkalization treatment, on the composite's thermal and mechanical properties. This data analysis aims to determine the optimal composition that provides the best properties for the rPP-based composite while contributing to the development of more environmentally friendly materials.

## 3. RESULT AND DISCUSSION

The product developed in this study is a composite based on recycled polypropylene (rPP) with the addition of Jatropha Seed Shell (JSS) as a filler, as shown in Figure 1. The research aimed to evaluate the effect of incorporating Jatropha Seed Shell (JSS) as a filler in rPP-based composites. Two methods were employed in this study: the first involved adding Jatropha Seed Shell (JSS) without any alkalization treatment, while the second used JSS treated with a 20% NaOH solution through an alkalization process.

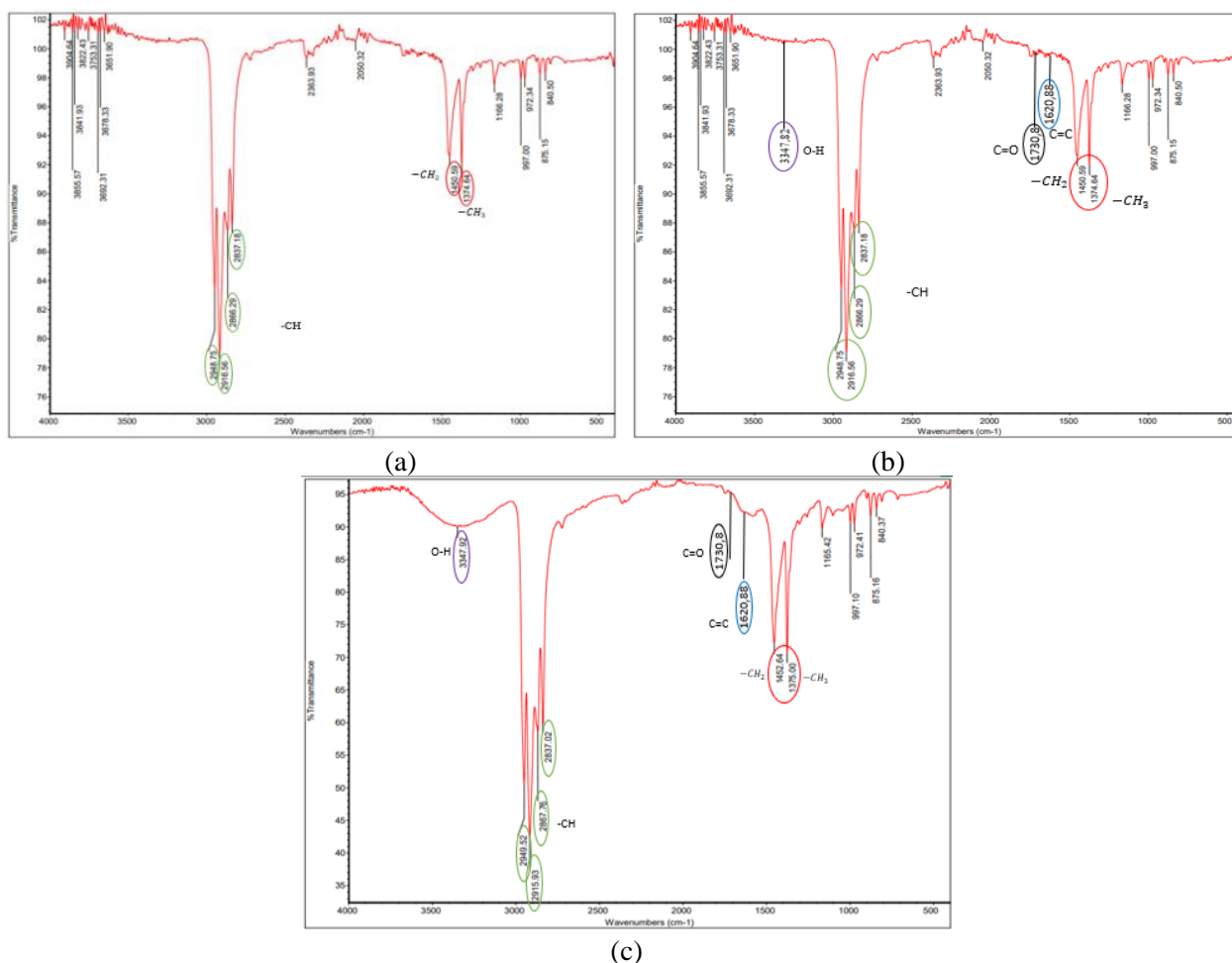
The method without alkalization treatment was designed to directly assess the impact of Jatropha Seed Shell (JSS) as a natural filler on the mechanical and thermal properties of the composite. Meanwhile, the alkalization treatment was intended to enhance the compatibility between JSS and the rPP matrix by removing components such as lignin and hemicellulose, which can reduce adhesion between the filler and the polymer [12] [13].



**Figure 2.** Recycled PP-Based Composite with Jatropha Seed Shell Filler

### 3.1. FTIR Testing

Functional group testing was conducted at the Testing Laboratory of UPPM, Department of Chemical Engineering, Universitas Indonesia. The analysis of the functional groups in Jatropha seed shells, both treated and untreated, was performed using Fourier Transform Infrared Spectroscopy (FTIR). The identification of functional groups is indicated by the shifting of absorption peaks or the appearance of new absorption peaks. The infrared spectra of 100% rPP, rPP/15% J Jatropha Seed Shell (JSS) without alkalization treatment, and rPP/15% JSS with alkalization treatment are shown in Figures 3a, 3b, and 3c.



**Figure 3.** Infrared spectra of 100% rPP (a), rPP/15% Jatropha Seed Shell (JSS) composite without alkalization treatment (b), and rPP/15% JSS composite with alkalization treatment (c)

Figure 3a illustrates the characteristics of recycled polypropylene (rPP) without filler, with the main functional groups being  $-CH$  ( $2950\text{ cm}^{-1}$  and  $2920\text{ cm}^{-1}$ ) and  $-CH_3$  ( $1375\text{ cm}^{-1}$  and  $1450\text{ cm}^{-1}$ ). The absence of oxygenated groups, such as carbonyl ( $C=O$ ), indicates that rPP is a non-polar polymer. Figure 3b shows the rPP/15% JSS composite without alkalization treatment, with the appearance of carbonyl ( $C=O$ ) bands at  $1730\text{ cm}^{-1}$  and hydroxyl ( $-OH$ ) bands at  $3348\text{ cm}^{-1}$ , originating from lignin and hemicellulose in Jatropha Seed Shell (JSS). This suggests imperfect filler distribution within the rPP matrix. Figure 3c reflects the rPP/15% JSS composite after alkalization treatment, with reduced intensity of carbonyl ( $C=O$ ) and hydroxyl ( $-OH$ ) bands, indicating partial removal of lignin and hemicellulose and an increase in hydrophobicity. The  $-CH$  groups remain dominant, reflecting the stability of the fundamental rPP structure. Alkalization treatment enhances compatibility between JSS and rPP, resulting in better filler distribution and improved composite quality [14], [15].

### 3.2. Melt Flow Rate Testing

The testing was conducted using a Melt Flow Indexer (MFI) in accordance with ASTM D1238 standards to determine the material's flow rate. This test measures the material flow rate in grams per 10 minutes, providing an indication of the composite's fluidity. It is also highly useful for assessing the viscosity of plastic materials when exposed to heat above their melting temperature. The effect of JSS composition on the Melt Flow Rate (MFR) is presented in Table 1.

**Table 1.** Effect of JSS Composition on MFR

Jatropha Seed Shell (JSS) (%)	MFR Value (g/10 minutes)	
	With Alkalization	Without alkalization
0	21.89	20.62
5	19.33	18.50
10	17.08	17.21
15	15.55	15.55
20	14.55	15.38

Table 1 presents the Melt Flow Rate (MFR) test results for recycled polypropylene (rPP)-based composites with the addition of Jatropha Seed Shell (JSS) as a filler, both with and without alkalization treatment. The MFR values decrease as the JSS percentage increases. For the composite without filler (0% JSS), the MFR value with alkalization treatment is 21.89 g/10 minutes, while without alkalization treatment, it is 20.62 g/10 minutes. As the JSS composition increases to 5%, 10%, 15%, and 20%, the MFR values for samples with alkalization treatment sequentially decrease to 19.33, 17.08, 15.55, and 14.55 g/10 minutes. For samples without alkalization treatment, the MFR values sequentially decrease to 18.50, 17.21, 15.55, and 15.38 g/10 minutes.

Overall, the results indicate that the MFR values of composites without alkalization are higher than those of composites with alkalization treatment at every percentage of JSS addition. The higher MFR values in composites without alkalization suggest that these materials exhibit greater fluidity, meaning lower viscosity compared to the alkalized composites [16]. The alkalization treatment is performed to remove lignin, hemicellulose, and other non-cellulosic components in JSS. This process makes the surface of JSS more reactive and more compatible with the rPP matrix. Consequently, the increased adhesion and compatibility between the JSS filler and the polymer matrix result in the alkalized composites being stiffer and less fluid, as reflected by the lower MFR values [17] [18] [19].

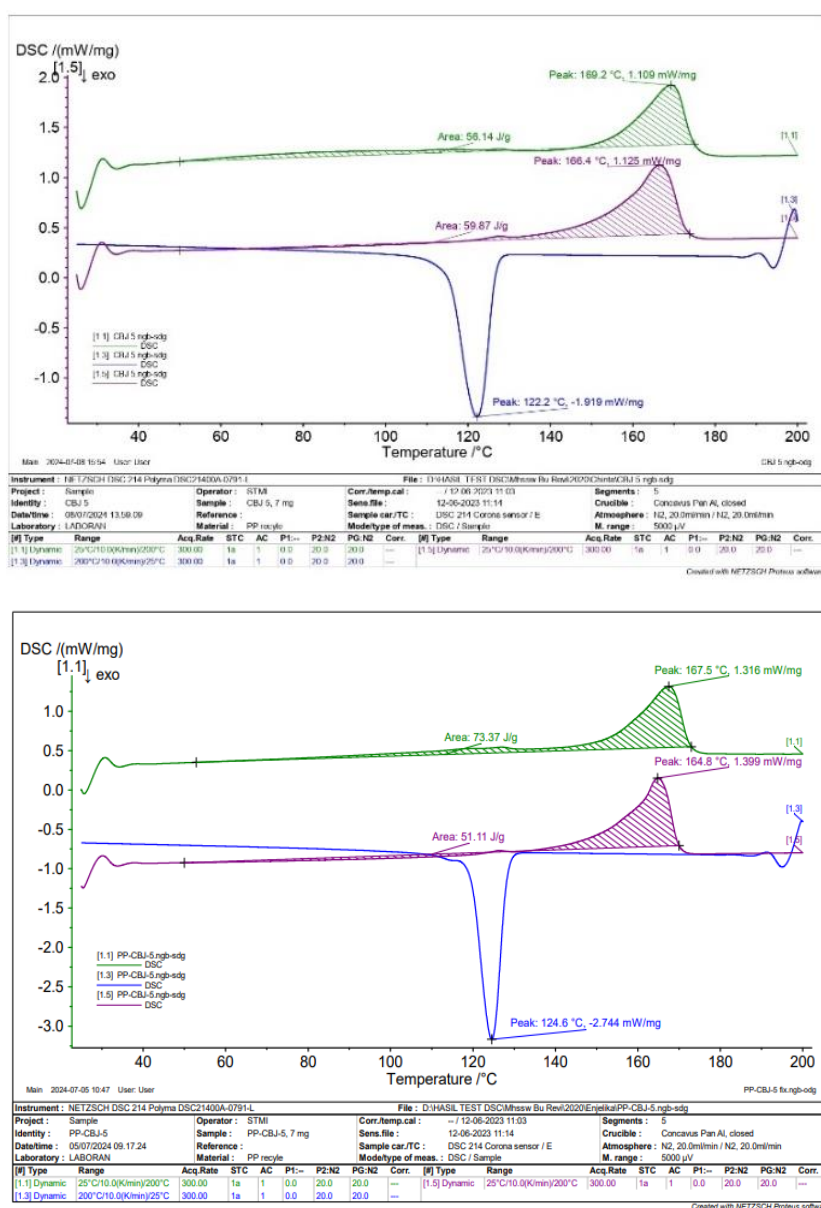
The lower MFR values in alkalized composites indicate higher viscosity, which is generally associated with improved mechanical properties such as tensile strength and stiffness [3]. As viscosity increases, the material becomes denser, enhancing its resistance to mechanical deformation. This makes alkalized composites more suitable for applications requiring higher mechanical strength, although their processability may become more challenging. On the other hand, composites without alkalization have higher MFR values,



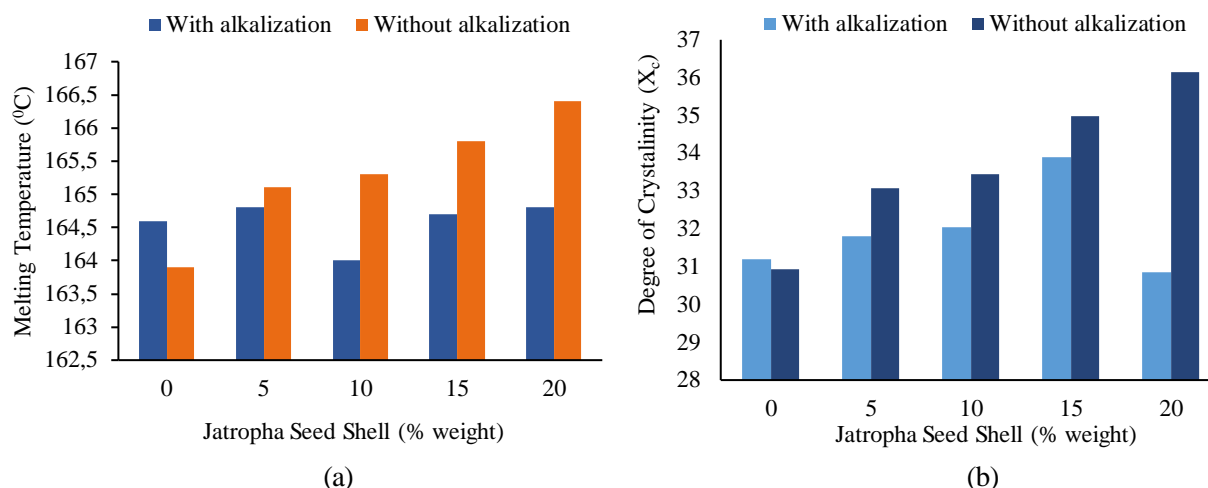
demonstrating better fluidity and lower viscosity. This suggests that these composites are easier to process using molding or extrusion techniques, but their mechanical properties may not be as robust as those of alkalized materials. Therefore, the addition of JSS filler, whether with or without alkalization treatment, should be tailored to the specific requirements of the composite application to achieve the desired balance between processability and mechanical performance [9].

### 3.3. Differential Scanning Calorimetry (DSC) Testing

Thermal testing using Differential Scanning Calorimetry (DSC) aims to evaluate the thermal properties of the composite, particularly related to the melting temperature ( $T_m$ ), enthalpy changes in the material, and the degree of crystallinity. The DSC thermogram results for composite materials with 20% by weight Jatropha Seed Shell (JSS) addition, both with and without alkalization treatment, are shown in Figures 4a and 4b. Meanwhile, the effect of JSS composition on melting temperature and degree of crystallinity is shown in Figures 5a and 5b.



**Figure 4.** Thermogram of recycled PP with 20% JSS addition with alkalization (a) and without alkalization (b)



**Figure 5.** Effect of Jatropha Seed Shell (JSS) composition on melting temperature (a) and degree of crystallinity (b)

In Figure 5a, it can be observed that the melting temperature ( $T_m$ ) of the recycled polypropylene (rPP)-based composite increases with the addition of Jatropha Seed Shell (JSS). Without JSS (0%), the melting temperature is  $163.9^{\circ}\text{C}$ . When JSS is added at 5%, 10%, 15%, and 20%, the melting temperature gradually increases to  $165.1^{\circ}\text{C}$ ,  $165.3^{\circ}\text{C}$ ,  $165.8^{\circ}\text{C}$ , and  $166.4^{\circ}\text{C}$ , respectively. This increase in melting temperature indicates that the addition of JSS filler enhances the thermal stability of the rPP composite. JSS, which contains cellulose, hemicellulose, and lignin acts as a reinforcing agent that improves the molecular structure's regularity within the polymer matrix, thereby strengthening the intermolecular bonds. This effect causes the composite to require a higher temperature to melt, reflecting an improvement in the material's thermal stability. The improved thermal stability also indicates a good interaction between Jatropha Seed Shell (JSS) and the rPP matrix, even without alkalization treatment. JSS contributes to enhancing the thermal stability of the composite. With alkalization treatment, the initial compositions (0-5% JSS) show a slight increase in  $T_m$ , but it tends to stabilize beyond 10%. At 20% JSS,  $T_m$  does not increase significantly compared to lower compositions. Alkalization treatment, which removes lignin and hemicellulose, improves the adhesion between JSS and the polymer matrix. However, the thermal stability effect of the alkalized filler seems to be less pronounced than that of the untreated filler, as the more uniform filler distribution reduces thermal resistance within the matrix [20].

Figure 5b illustrates the effect of Jatropha Seed Shell (JSS) composition on the degree of crystallinity ( $X_c$ ) of the rPP composite. The results indicate a significant difference between JSS with and without alkalization treatment. In samples without alkalization treatment, the degree of crystallinity increases as the percentage of JSS increases. With the addition of JSS at 5%, 10%, 15%, and 20%, the degree of crystallinity gradually rises to 33.07%, 33.45%, 34.97%, and reaches the highest value at 36.14%. The increase in  $X_c$  suggests that untreated JSS acts as a crystallization agent that enhances molecular regularity within the polymer matrix. Without alkalization treatment, Jatropha Seed Shell (JSS) filler may serve as an effective nucleation centers, aiding in the formation of more crystalline regions [21].

With alkalization treatment, the degree of crystallinity tends to be lower compared to untreated samples. With the addition of JSS at 5%, 10%, and 15%, the degree of crystallinity gradually increases to 31.81%, 32.04%, and peaks at 33.90% for the 15% JSS composition. However, at 20% JSS, the degree of crystallinity decreases to 30.85%. This indicates that at higher compositions (20%), alkalization treatment may result in more uniform filler distribution but reduces JSS's ability to act as a nucleating agent for crystallization. While alkalization improves filler adhesion, it may reduce the number of free nucleation sites available on the surface of JSS. This diminishes JSS's potential as a crystallization agent that could enhance the degree of crystallinity.

Untreated JSS still contains lignin and hemicellulose, which may act as physical barriers within the polymer matrix. These barriers could promote the formation of more nuclei or crystallization centers, thereby increasing the degree of crystallinity. In contrast, alkalized JSS has a cleaner surface, reducing the number of potential nucleation sites, which in turn decreases its ability to enhance the degree of crystallinity [22].

### 3.4. Density Testing

Based on the density measurements of rPP without JSS addition and with 15% JSS addition, a decrease in density was observed from 0.847 g/mL to 0.718 g/mL. This reduction can be interpreted as a direct effect of adding JSS filler, which has a lower density compared to the rPP polymer matrix. When JSS is added, it replaces part of the denser polymer with a lighter material, resulting in a lower mass per unit volume of the composite and thus reducing the overall density of the composite. These results are consistent with previous studies, which show that the addition of low-density fillers to a polymer matrix tends to reduce the overall density of the composite. For example, a study by Ilori [23] reported that the incorporation of lightweight particle-based fillers, such as fly ash and natural fibers, significantly reduced the density of polymer composites due to the lightweight nature of the fillers. Another study by Dubey demonstrated that the use of shell powder as a filler in recycled polypropylene also reduced the composite's density because of the material replacement effect with lightweight fillers [24]. Moreover, Dwivedi found that partially replacing the polymer matrix with organic waste-based fillers could enhance the weight efficiency of the composite without compromising certain mechanical properties, supporting the findings that low-density fillers contribute to a reduction in the overall density of the material [25].

Additionally, the inclusion of JSS may create voids or pores within the composite. JSS may not be evenly distributed within the matrix, or there may be imperfect adhesion between JSS and the polymer matrix. These voids reduce the overall material density due to the presence of empty spaces within the composite. The addition of JSS can also lead to uneven distribution of filler particles within the matrix. If the particles are not uniformly dispersed, areas with low and high filler concentrations may form, resulting in local variations in material density and ultimately causing a decrease in the composite's overall density [26] [27].

## 4. CONCLUSION

The addition of Jatropha Seed Shell (JSS) to rPP, both with and without alkalization treatment, resulted in a decrease in MFR values as the JSS composition increased. The MFR dropped from 21.89 g/10 minutes (0% JSS) to 14.55 g/10 minutes (20% JSS) with alkalization treatment, and from 20.62 g/10 minutes to 15.38 g/10 minutes without alkalization. This decline indicates an increase in viscosity, particularly for alkalized JSS, due to improved adhesion between the filler and the matrix. The effect of adding JSS was also evident in the increase in the composite's melting temperature ( $T_m$ ). The study showed that the  $T_m$  of the composite increased from 163.9°C (0% JSS) to 166.4°C (20% JSS) without alkalization treatment, while with alkalization treatment,  $T_m$  remained relatively stable in the range of 164.0°C to 164.8°C. The increase in  $T_m$  indicates that JSS acts as a reinforcement, enhancing the thermal stability of the composite, thereby requiring higher temperatures to reach the melting point. However, composites with untreated JSS exhibited a more significant increase in  $T_m$ , suggesting that untreated JSS is more effective in facilitating the formation of a more stable crystalline structure compared to alkalized JSS. The degree of crystallinity ( $X_c$ ) also showed an increasing trend with higher JSS composition. Without alkalization treatment,  $X_c$  increased from 30.92% (0% JSS) to 36.14% (20% JSS), which was more pronounced compared to alkalized samples, where  $X_c$  increased from 31.20% (0% JSS) to 33.90% (15% JSS). Although alkalization treatment improves filler compatibility, it tends to reduce JSS's ability to act as a crystallization agent due to a decrease in the number of free nucleation sites on the JSS surface. The reduction in density from 0.847 g/mL to 0.718 g/mL after the addition of 15% JSS indicates that the filler reduces the composite's mass per unit volume. This reduction is attributed to the difference in density between JSS and the polymer matrix, the potential formation of pores within the composite structure, and possible imperfections in the distribution of the filler within the matrix.



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