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SCUOLA DI INGEGNERIA INDUSTRIALE E DELL'INFORMAZIONE

Executive Summary of the Thesis

Manufacturing and characterization of fiber reinforced polymers with natural fibers and recyclable matrix

Laurea Magistrale in Materials Engineering and Nanotechnology - Ingegneria dei Materiali e delle Nanotecnologie

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1. Introduction

The growing demand for more sustainable engineering materials has increased interest in fiber-reinforced polymers based on natural reinforcements and recyclable matrices. In this context, natural fiber-reinforced composites (NFRCs) have emerged as a promising alternative to conventional composites reinforced with synthetic fibers, since they can offer lower density, reduced environmental impact, and good specific mechanical properties for semi-structural applications [1, 2].

Among thermoplastic matrices, polypropylene (PP) is particularly attractive because of its low density, low cost, and compatibility with established industrial processes such as extrusion and injection molding. However, the performance of PP-based natural fiber composites remains strongly dependent on fiber dispersion, interfacial adhesion, moisture sensitivity, and microstructural variability driven by processing conditions. In addition, the effects of hybridization and reprocessing are not yet fully clarified, especially when different fiber systems are compared under the same manufacturing and testing conditions.

Within this framework, this thesis investigates polypropylene-based composites reinforced with pineapple fibers (PF), jute fibers (JF), and their hybrid PF/JF combinations, considering both virgin and mechanically reprocessed materials. The objective is to provide a consistent experimental comparison of these systems and to assess how fiber type, fiber content, hybridization, and reprocessing affect consolidation quality, physical behavior, and mechanical performance. To this end, the study is based on melt compounding in a twin-screw extruder followed by injection molding, and on a characterization campaign including density measurements, tensile, flexural, and impact testing, water uptake experiments, and SEM observations of fracture surfaces.

2. Materials and methodology

The investigated systems were polypropylene (PP)-based composites reinforced with pineapple fibers (PF), jute fibers (JF), and PF/JF hybrid combinations, with neat PP used as the baseline reference. The composite set included both single fiber and hybrid formulations at two reinforcement levels (15 and 30 wt%). In addi-

Material	Family	Density	Tensile	Flexural	Impact	Water uptake
PP	virgin	✓	✓	–	✓	–
PF15(1)	virgin	✓	✓	✓	✓	✓
PF15(2)	virgin	✓	✓	✓	✓	–
PF30	virgin	✓	✓	✓	✓	✓
JF15	virgin	✓	✓	✓	✓	✓
JF30	virgin	✓	✓	✓	✓	✓
HYB15	virgin	✓	✓	✓	✓	–
HYB30	virgin	✓	✓	✓	✓	–
PPR	recycled	✓	✓	–	✓	–
PF15(1)R	recycled	✓	–	–	–	–
PF15(2)R	recycled	✓	✓	–	✓	–
PF30R	recycled	✓	✓	–	✓	–
JF15R	recycled	✓	✓	–	✓	–
JF30R	recycled	✓	✓	–	✓	–
HYB15R	recycled	✓	✓	–	✓	–
HYB30R	recycled	✓	✓	–	✓	–

Table 1: Summary of the investigated formulations and corresponding experimental datasets.

tion, for selected systems, mechanically reprocessed (recycled) counterparts were produced to evaluate the effect of reprocessing on consolidation and final properties. A summary of the investigated formulations and corresponding experimental datasets is reported in Table 1. The material ID in the left column of Table 1 shows the type of fiber and its content in the composite (e.g. JF15 is PP reinforced with 15 wt.% jute fibers, while HYB30 shows a hybrid reinforcement content of 30 wt.%). The "R" suffix indicates a recycled composite.

The materials were produced by melt compounding in a twin-screw extruder followed by injection molding of the test specimens. After the first processing cycle, selected materials were reprocessed and injection molded again to quantify the impact of reprocessing on consolidation and performance.

Characterization combined physical, mechanical, and microstructural analyses (see Table 1). Density measurements were used to compare experimental and theoretical values as an indirect indicator of consolidation quality. Mechanical performance was evaluated through tensile (ISO 527), flexural (ISO 178), and impact (ISO 180) tests. Moisture sensitivity was assessed via water uptake at different temperatures, and SEM observations of fracture surfaces were used to support the interpretation of the mechanical results.

3. Results and discussion

The results of this study are presented by focusing on the most representative indicators of processing quality, mechanical performance, and microstructural integrity. In particular, the discussion is centered on density, tensile modulus, tensile strength, and impact toughness, since these parameters most effectively capture the main effects of fiber type, fiber content, hybridization, and reprocessing. Density was selected as an indirect indicator of consolidation quality, tensile modulus to highlight the stiffening effect of the reinforcement, tensile strength to quantify the maximum stress sustained under quasi-static loading, and impact toughness to assess the loss of fracture resistance after reprocessing. These results are further supported by water-uptake measurements and SEM observations, which help relate macroscopic behavior to the underlying microstructure.

3.1. Processing quality

Density measurements proved to be a useful first indicator of consolidation quality. As shown in Figure 1, deviations between experimental and theoretical density were associated with differences in impregnation efficiency and void content, with larger discrepancies generally corresponding to poorer consolidation. This effect became more relevant at higher fiber content, where the increase in melt viscosity made wetting and compaction more difficult. In contrast, the recycled composites systematically exhibited higher density than their virgin counterparts, indicating a clear densification effect after reprocessing and suggesting a reduction in macroscopic porosity. This trend was especially evident for the 30 wt% single-fiber sys-

tems, whereas it was more limited for the 15 wt% formulations and for the hybrid materials, which already showed relatively good consolidation in the virgin state.
An additional result concerns the comparison between PF15(1) and PF15(2), which highlighted the strong influence of compounding strategy. Although both formulations had the same nominal composition, they showed different density levels and distinct fracture surface features, confirming that feeding strategy and mixing efficiency can significantly affect the final microstructure.

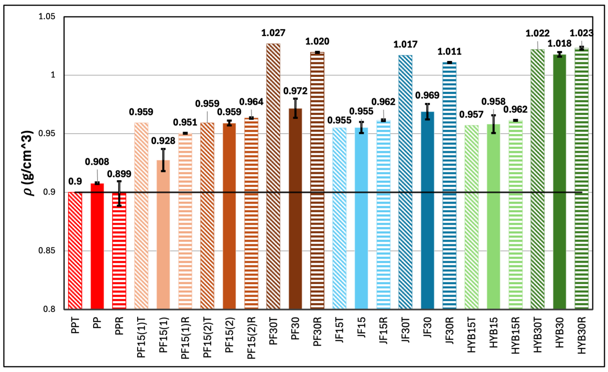

Figure 1: Experimental and theoretical density of neat PP, virgin composites, and recycled composites.

3.2. Tensile behavior

The tensile results highlighted a clear reinforcement effect relative to neat PP, together with marked differences among fiber families and processing conditions. As shown in Figure 2, JF-based composites tended to provide slightly higher stiffness than PF systems at the same fiber content, indicating a more effective elastic contribution under the adopted processing conditions. Conversely, PF-based systems reached higher tensile strength at both 15 and 30 wt% (Figure 3), suggesting a more favorable balance between reinforcement efficiency and resistance to premature crack initiation in the present dataset.
Increasing fiber content from 15 to 30 wt% produced a consistent stiffening effect and increased density, confirming that the elastic response became progressively more influenced by the reinforcement. However, the higher reinforcement level also made the composites more sensitive to defects and local stress concentrations, especially in terms of strength and strain at break.

Hybridization provided an intermediate and, in some cases, well-balanced response. In particular, at 30 wt%, the hybrid system showed competitive tensile behavior, indicating that combining PF and JF can promote effective stress redistribution when the microstructure is sufficiently homogeneous.
The comparison between virgin and recycled composites showed that reprocessing did not produce a uniform effect on tensile performance. In general, recycled materials often retained, and in some cases slightly increased, tensile modulus (Figure 2), consistently with the densification observed in the density measurements. By contrast, tensile strength did not show the same systematic improvement (Figure 3), suggesting that while improved consolidation can contribute positively to stiffness, local fiber damage, interfacial modifications, and matrix degradation may limit the preservation of strength [3].

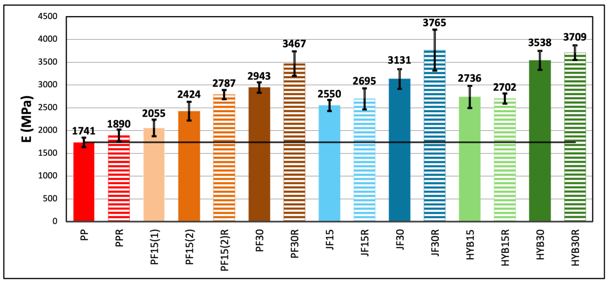

Figure 2: Tensile modulus of neat PP, virgin composites, and recycled composites.

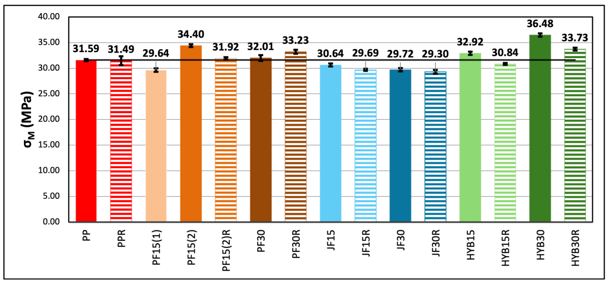

Figure 3: Tensile strength of neat PP, virgin composites, and recycled composites.

3.3. Impact behavior

In contrast to tensile behavior, the impact results revealed a different response across the investigated formulations. As shown in Figure 4, the 30 wt% composites generally outperformed the corresponding 15 wt% systems, indicating that the higher reinforcement level could en-

hance impact resistance under the tested conditions. Among all the materials, PF30 showed the highest impact toughness, suggesting that the PF-based system provided the most favorable balance between reinforcement and energy absorption in the present dataset.
The hybrid composites exhibited an intermediate response. Although hybridization led to a balanced overall behavior, the impact performance of the hybrid systems did not exceed that of the best PF-based formulation. This suggests that, under the adopted processing and testing conditions, combining PF and JF did not produce a marked advantage in notch-impact resistance.
The comparison between virgin and recycled composites showed a much more consistent trend than in tensile loading. After reprocessing, impact toughness decreased for every recycled formulation, even when density increased. This indicates that the densification observed after reprocessing was not sufficient to preserve fracture resistance under dynamic loading. Rather, the results suggest that impact performance after recycling was governed by additional damage mechanisms, such as fiber shortening, local interfacial modifications, and matrix embrittlement [3].
Overall, these findings show that reprocessing improved consolidation but simultaneously reduced the ability of the material to absorb impact energy. As a result, impact toughness emerged as the most sensitive property for detecting the negative effects of recycling in the investigated composites.

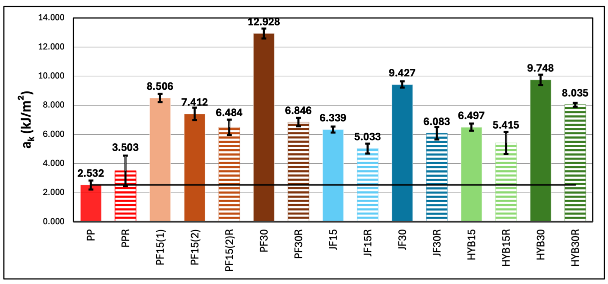

Figure 4: Izod impact toughness of neat PP, virgin composites, and recycled composites.

3.4. Water uptake and microstructure

Water uptake measurements and SEM observations were used to complement the mechanical results by providing additional insight into moisture sensitivity and fracture surface morphology. In particular, the water uptake curves shown in Figure 5 help clarify the role of fiber content and temperature on moisture sensitivity, while the SEM comparison in Figure 6 supports the interpretation of the main microstructural trends.
The water uptake results confirmed the expected sensitivity of natural fiber-reinforced systems to moisture exposure [4]. For both PF- and JF-based composites, increasing fiber content led to higher long term mass uptake, consistent with the greater fraction of hydrophilic reinforcement and the higher probability of interfacial pathways for moisture transport. As expected, increasing the immersion temperature from 20 °C to 60 °C accelerated the absorption kinetics and amplified the differences among formulations, making the role of fiber type and content more evident.
At 20 °C, the overall trend remained more gradual, with the 30 wt% formulations showing higher long term uptake than the corresponding 15 wt% systems. At 60 °C, the separation among the curves became clearer, confirming that elevated temperature enhances moisture diffusion and increases the sensitivity of the response to local defects and interfacial discontinuities. Overall, these results are consistent with the hydrophilic nature of the fibers and with the role of processing quality in controlling the accessibility of moisture to the composite interior.

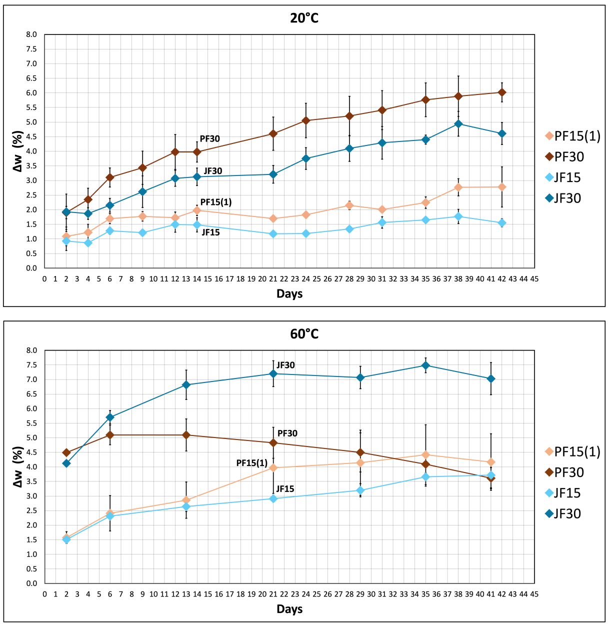

Figure 5: Water uptake as a function of immersion time for the investigated composite formulations at 20 °C (top) and 60 °C (bottom).

SEM observations further supported the interpretation of the physical and mechanical results. As shown in Figure 6, only the JF-based composites are reported here as a representative example; however, similar trends were also observed for the PF-based systems. In particular, the comparison between virgin and recycled JF composites highlighted a reduction in macroscopic voids after reprocessing, in agreement with the density increase measured for the recycled materials. By contrast, for the hybrid systems the differences between virgin and recycled conditions appeared less pronounced, consistently with the smaller density variations already discussed in the previous section.
At the same time, the SEM evidence suggests that improved densification did not necessarily lead to better overall performance, since the reduction in impact toughness after recycling points to additional microstructural changes, such as fiber damage or interfacial modifications.

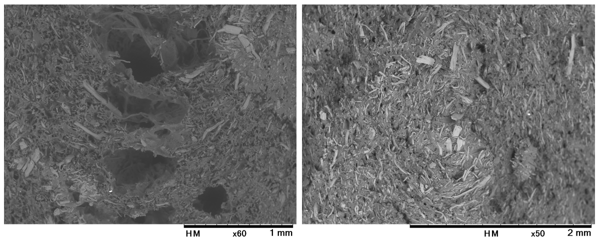

Figure 6: SEM micrographs of tensile fracture surfaces comparing virgin (left) and recycled (right) JF-based composites at low magnification (30 wt% fiber content).

Overall, the water uptake and SEM results confirm that the behavior of the investigated composites was governed not only by composition, but also by the effects of processing history on consolidation, moisture transport, and fracture mechanisms.

4. Conclusions

This work provided a consistent experimental assessment of polypropylene-based composites reinforced with pineapple fibers, jute fibers, and their hybrid combinations, considering both virgin and mechanically reprocessed materials. Overall, the results showed that the final behavior of these systems is governed not only by nominal composition, but also by the strong interaction between processing quality, microstructural integrity, and reinforcement architecture. This confirms the importance of controlling dispersion, consolidation, and interfacial quality when designing PP-based natural fiber composites, in agreement with the trends reported for similar PF- and JF-reinforced systems [5].
Overall, these results support the feasibility of using PF-, JF-, and hybrid-reinforced PP composites as promising lightweight and more sustainable material solutions, while also showing that recycling strategies must be carefully optimized to preserve damage tolerance and long term performance.

References

[1] Omar Faruk, Andrzej K. Bledzki, Hans-Peter Fink, and Mohini Sain. Biocomposites reinforced with natural fibers: 2000–2010. *Progress in Polymer Science*, 37:1552–1596, 2012. doi: 10.1016/j.progpolymsci.2012.04.003.

[2] Barshan Dev, Md. Ashikur Rahman, Md. Rubel Alam, Md. Reazuddin Repon, and Yasir Nawab. Mapping the progress in natural fiber reinforced composites: Preparation, mechanical properties, and applications. *Polymer Composites*, 44: 3748–3788, 2023. doi: 10.1002/pc.27376.

[3] M. Zampaloni, F. Pourboghrat, S.A. Yankovich, B.N. Rodgers, J. Moore, L.T. Drzal, A.K. Mohanty, and M. Misra. Kenaf natural fiber reinforced polypropylene composites: A discussion on manufacturing problems and solutions. *Composites Part A: Applied Science and Manufacturing*, 38:1569–1580, 2007. doi: 10.1016/j.compositesa.2007.01.001.

[4] M. Mohammed, A. J. A. M. Jawad, A. M. Mohammed, J. K. Oleiwi, T. Adam, A. F. Osman, and M. Jaafar. Challenges and advancement in water absorption of natural fiber-reinforced polymer composites. *Polymer Testing*, 124:108083, 2023. doi: 10.1016/j.polymertesting.2023.108083.

[5] M. M. Alamgir Sayeed, Abu Sadat Muhammad Sayem, Julfikar Haider, Sharmin Akter, Md. Mahmudul Habib, Habibur Rahman, and Sweety Shahinur. Assessing mechanical properties of jute, kenaf, and pineapple leaf fiber-reinforced polypropylene composites: Experiment and modelling. *Polymers*, 15:830, 2023. doi: 10.3390/polym15040830.