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New Trends in Polymer Science

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Universidad de los Andes Colombia

# SUSTAINABLE 3D PRINTING FILAMENTS BASED ON RECYCLED POLYPROPYLENE AND RICE HUSK: EFFECTS OF FIBER PARTICLE SIZE AND MALEIC ANHYDRIDE-GRAFTED-POLYPROPYLENE IMPLEMENTATION

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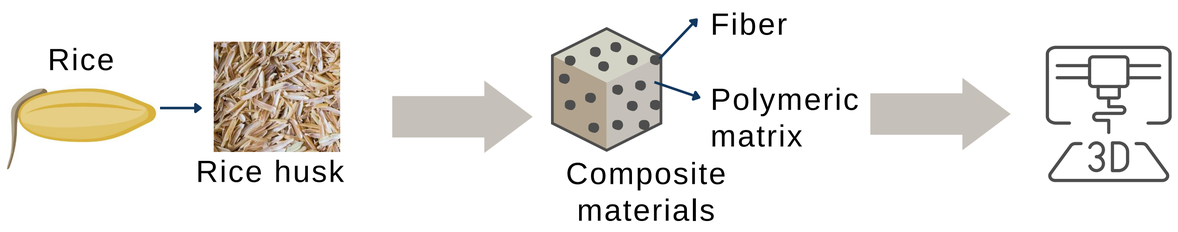
## ABSTRACT

Nowadays, scientists and industry are concerned with protecting the planet, by enhancing the development of more sustainable materials. In addition, the development of additive manufacturing techniques enables locally produced parts with minimal waste and would benefit from the use of renewable raw materials. This work presents the development and characterization of 3D printing filaments based on recycled polypropylene (rPP) and rice husk (RH). The influence of two different particle sizes and the implementation of maleic anhydride-grafted-polypropylene (MAPP) as an additive on the 3D printing process and 3D printed parts were evaluated. Warping effect, thermal, tensile, and morphological analyses were performed for the study. Results showed an improvement in tensile properties and reduction of warping when MAPP was implemented together with the use of smaller particle size of the RH. According to the SEM analysis, this behavior was caused by an improved interfacial adhesion between the polymeric matrix and the fiber achieved with the MAPP. A better fiber distribution is achieved when smaller particle size is used. The study demonstrates the potential of recycled polymers and agro-industrial waste in new manufacturing techniques, like 3D printing, contributing to the change to a circular economy industrial model.

## INTRODUCTION

In Colombia, around 72 million tons of residual agricultural biomass is produced each year by different industries [1], such as the rice industry, which is responsible for making 700 thousand tons of RH [2,3]. In addition, plastic pollution is one of the most significant environmental threats nature and humans face due to its accumulation in natural resources. PP is one of the most commonly found due to its multiple applications. Currently, solid waste as raw material for other industries has gained interest to reduce environmental pollution and minimize the use of natural renewable and non-renewable resources.



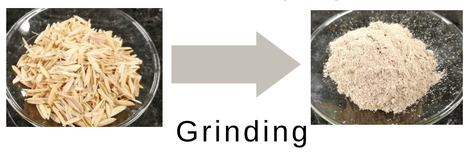

Recently, the development of more sustainable technologies has also gained importance. For example, Fused Deposition Modeling, an additive manufacturing technology, has been increasing in the last years due to its versatility in manufacturing new products, conceptual designs, functional parts, and tooling [4]. An alternative to the common polymeric materials is natural fiber composite materials. Natural fibers are renewable, non-toxic, compostable, recyclable, and abundant materials. In the 3D printing of composite materials, thermal, mechanical, and morphological properties can be influenced by parameters such as natural fiber particle size and the implementation of additives. MAPP is an additive frequently used in polypropylene composites to enhance the interaction between natural fibers and PP matrix [5]. Thus, this study evaluates the influence of two different fiber particle sizes and MAPP implementation on 3D printed parts over their thermal, mechanical, and morphological properties.

## METHODS

1. Materials

rPP RH MAPP

2. Raw materials preparation



Grinding
Sieving

Sieve mesh
40-60 (L): 250 - 425 µm
80-100 (S): 150 -180 µm

3. 3D printing filament development

Pellets

| Run | MAPP | Particle |
|---|---|---|
| 0 | No | - |
| 1 | Yes | Small |
| 2 | No | Small |
| 3 | Yes | Large |
| 4 | No | Large |

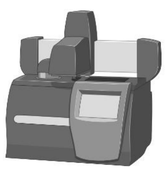
Filament

Temperature profile 175 - 190°C

Temperature profile 175 - 190°C

4. Thermal characterizarion

Differential Scanning Calorimetry ASTM D3418



5. Specimens 3D printing

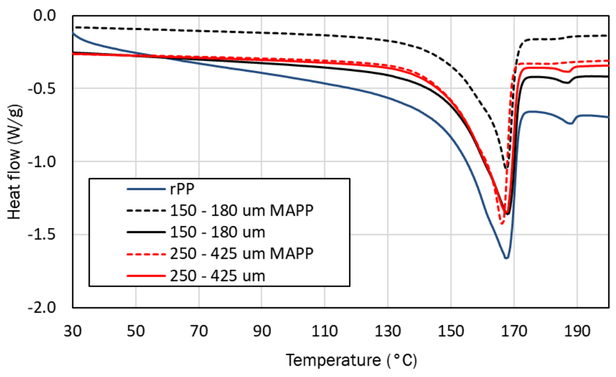
ASTM 3039
Extruder - 250°C
Bed - 80°C
Build surface - PP tape

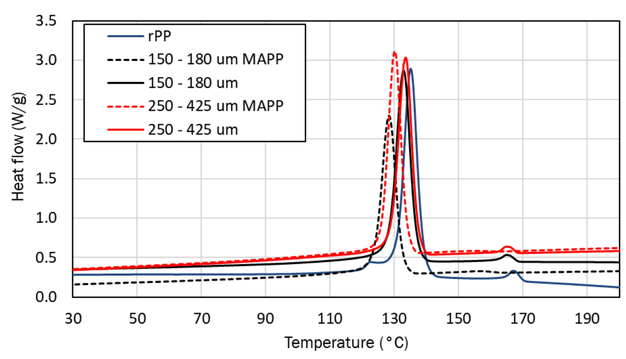
6. Characterization

- Warping effect
- Tensile test D3039
- Scanning Electron Microscopy

## RESULTS

1. Thermal characterization - DSC results

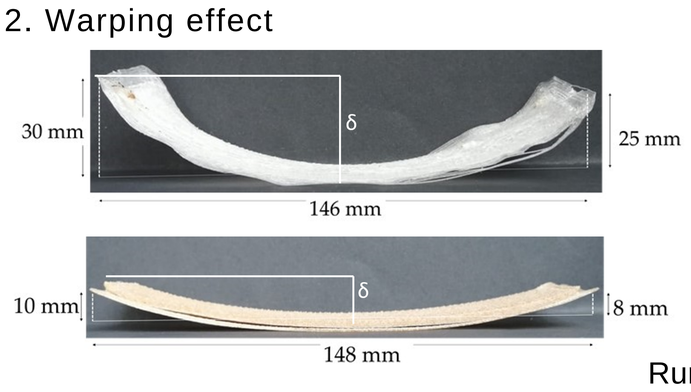



| Run | Tc (°C) | dHc (J/g) | Tm (°C) | dHm (J/g) | Crystallinity (%) |
|---|---|---|---|---|---|
| 0 | 135.1 | 84.3 | 167.3 | 94.8 | 45.8 |
| 1 | 128.3 | 70.9 | 167.6 | 64.1 | 34.4 |
| 2 | 132.9 | 80.1 | 167.8 | 85.4 | 45.8 |
| 3 | 130.2 | 85.4 | 166.1 | 74.9 | 40.2 |
| 4 | 133.5 | 85.7 | 167.4 | 83.2 | 44.7 |

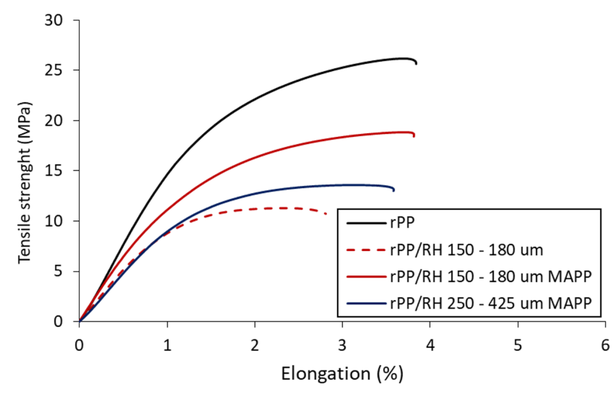
- MAPP causes a decrease in the material's crystallinity.
- A reduction in crystallization could improve the rPP 3D printing process.
- A small peak was perceived, which can be attributed to impurities due to its recycled nature.

2. Warping effect




- The warping effect is improved by 62% with MAPP implementation and smaller particle size.

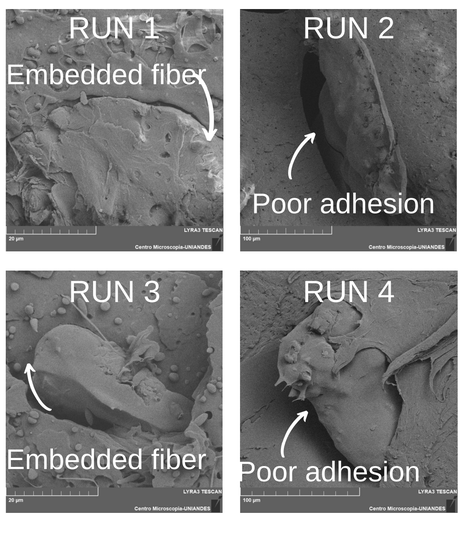
3. Tensile properties

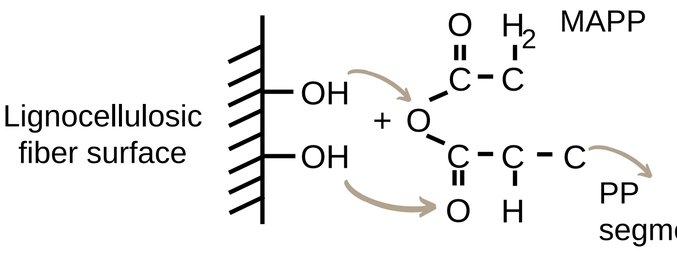



| Run | Tensile strength (MPa) | Tensile elongation (%)* | Young modulus (GPa) |
|---|---|---|---|
| 0 | 24.4 ± 2.0 | 24.4 ± 2.0 | 1.4 ± 0.2 |
| 1 | 20.4 ± 4.8 | 20.4 ± 4.8 | 1.4 ± 0.2 |
| 2 | 11.7 ± 2.1 | 11.7 ± 2.1 | 1.0 ± 0.1 |
| 3 | 14.8 ± 2.5 | 14.8 ± 2.5 | 1.0 ± 0.2 |
| 4 | - | - | - |

- rPP/RH with big particle size 3D printing filament could not be printed into specimens. Material blocked the nozzle and not homogeneous diameter was obtained.
- Tensile properties of specimens manufactured with small particle fiber size and anhydride is comparable to the rPP.

3. SEM graphs




- MAPP provides polar interactions and covalent links with the hydroxyl groups present in lignocellulosic fibers [6].
- MAPP addition improves the interfacial adhesion between the and RH.
- A smaller particle size allows a better dispersion of the fiber in the composite.

## CONCLUSIONS

Improvement in tensile properties and reduction of warping effect was achieved with MAPP implementation and the use of smaller particle size of the RH. This improvement was caused by tMAPP's link effect between RH fiber and rPP. A better fiber distribution was achieved when a smaller particle size was used. The study demonstrates the potential of recycled polymers and agroindustrial waste to be used in 3D printing.

## ACKNOWLEDGMENT

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