Mechanical Property Enhancement of Polypropylene Through Sustainable Epoxy Doping Employing Advanced Additive Manufacturing Technique

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Abstract: *This paper characterizes the mechanical properties and morphology of polypropylene (PP) and sustainable epoxy (SE) composites before and after the UV degradation experiment. PP and SE polymer composites were prepared through melt blending and were used to prepare 3D-printed samples. The obtained specimens were exposed to UV radiation for up to 3000 hours, and the tensile properties and surface microstructures were characterised through SEM. This study shows that the sample's tensile properties are decreased by UV irradiation due to PP photodegradation and that adding 1-5 wt % SE increases tensile strength and opposes UV impact. Compared with the materials under test the extruded PP with SE has the highest young modulus and strength while the Filament PP has the poorest mechanical properties. Results indicated the composite containing 1% SE has the highest tensile strength while the one having 2% SE has the highest young modulus. This work highlights the potential of SE-doped PP as a cost-effective and environmentally sustainable material for additive manufacturing, offering improved UV stability for diverse applications.*

Keywords: Polypropylene; Extrusion; 3D Printing; Mechanical Properties

1. INTRODUCTION

According to the literature review of 3D printing methods of polymers [1], Fused Deposition Modelling (FDM) is widely used and considered the most advanced technique in the global manufacturing system [2,3] due to its simplicity and low-cost invention and tool. Also referred to as Fused Filament Fabrication (FFF) due to the absence of definitive industry standards in additive manufacturing [4], this method helps to print a figure one layer after another layer, extruding the specified material through a nozzle supplied with a suitable filament. In most cases, the only constraint in this kind of technique is just the narrow range of materials that are part of the additive manufacturing materials list [5–7]. The available FDM common polymers on the market are limited to include "Polylactic acid (PLA), Nylon, Acrylonitrile butadiene styrene (ABS), or polycarbonate (PC)", FDM with almost no specific functionalization. The past several years has seen an abundance of this in the realm of science, with some studies being conducted to examine the behavior of various "better-performing polymer-based composites, [8,9] reinforced with natural and synthetic fillers [10-12]. Throughout the improvement process, Duty et al. attempted to develop the most appropriate model to evaluate polymers as extrusion materials for the intended Additive Manufacturing PM, however, there are certain requirements that a material must meet both during the extrusion process and adhering to the deposition of the extruded material. [13]. However, the issue here is that there is still no sufficient understanding of what particular characteristics of a material will ensure it is 3D printable, and thus, most of the knowledge is hidden and proprietary by the industrial stakeholders. For the past three decades, the "market of 3D printing technologies has expanded and extended to many different industries such as biomedical, automotive, aerospace, and so on, and now its primary objective is to take it to building, electronics, and designing." [14,15].

Applying this benefit, the greatest strength of additive manufacturing is its liberty from the final shape of the product: this shape is fully customizable with virtually zero costs, and as a result, the largest possible variety of materials is required to hype up this technology to the next level. [16,17].

By the nature of their processing, polymers can be grouped into the following categories: elastomer, thermoplastic, and thermosets. Elastomers are described as substances that if once stretched it can revert to their original shape at least twice as large as they were at room temperature. Thermoplastics are plastics that can be melted and remolded into different shapes without experiencing any significant changes in their characteristics. On the other hand, a thermoset may be described as plastic that cannot be reheated or reshaped into another form. Besides, virgin or origin petroleum based on the polymer are in many types where some of them are polypropylene pp, "polyvinyl chloride PVC, low-density polyethene LDPE, high-density polyethene hdpe", polyethene PE, and polystyrene PS.

Polypropylene (PP) has become a favourite among all thermoplastic polymers because of its advantageous characteristics, moderate price, and workability [18]. In the years that followed, serious efforts went into growing PP as well as in its application resulting in being used in a multitude of operations across the spectrum [18]. The fact that PP is so useful stems from its molecular structure of propylene monomers linked in a straight runway. This linear configuration suggests several appealing characteristics; higher crystallinity, better chemical

durability, lower density, and adequate mechanical properties [19]. These features qualify PP for several uses; in the manufacturing of packaging materials, customer products, auto parts, and medical devices [20]. In recent years, researchers have been studying the factors and ways of enhancing the features of PP, by creating copolymers, modifying with additives, or making nanocomposite [19,21]. PP is a highly significant polymer in the global economy, and its integration into additive manufacturing (AM) could lead to transformative changes. While the machinery properties of "3D printing grade" PP feedstocks have been thoroughly investigated in connection with parameters related to printing, practical considerations remain underexplored [22]. The fused filament fabrication (FFF) process introduces a complex thermal history that alters the crystallization kinetics of PP, leading to considerable volumetric shrinkage in printed parts [23]. This cooling-induced shrinkage traps residual stresses within the printed objects. The situation is exacerbated in PP due to its high crystallinity resulting from the regular arrangement of molecular chains [24]. One common solution to mitigate warpage is the incorporation of high aspect ratio fillers, such as carbon fibres. These fibres enhance thermal conductivity, promoting a more uniform temperature distribution within the print, which helps reduce internal stresses. Additionally, when aligned with the extrusion direction, the stiff fibres help resist warping during printing. However, the presence of fillers can hinder interlayer polymer chain diffusion, resulting in significantly weaker mechanical properties when tested perpendicular to the fibre orientation [25]. To address the challenges of warpage and interlayer strength without using fibres, blending PP with other polymers has emerged as an interesting alternative, demonstrating some success. PP composites reinforced with sustainable epoxy (SE) show that it improves mechanical strength in tensile tests.

Sustainable epoxy (SE) includes “palm oil, rapeseed oil, soybean oil, sunflower oil, and other types of epoxies”. These polymers mostly are made up of triacylglycerols, which are generated by the amination of fatty acid with glycerine. About 95% of the weight of triglycerides includes fatty acids, and the fatty acid profile differs in every type of waste oil. On the same note, “starch, lignin, protein, cellulose, chitosan, shellac, resin, polyhydroxyalkanoates furanone, alginate wool fibres, and waste vegetable oil” have been incorporated into polymer synthesis to reveal an increasing list of sustainable materials. However, epoxy adhesives are examples of the most widely used structural adhesives across the aerospace as well as automotive industries because of the best adhesion besides corrosion. The most renewable resources of feedstock are vegetable oils which have gained much attention for use in the synthesis of bio-based polymers due to their availability, biodegradability, and cheaper. Vegetable oils in the epoxy form have been widely used in the development of different epoxy adhesives that have good bonding characteristics on a large range of substrates such as “glass, steel, bamboo, and wood veneer”. The properties which include the mechanical and physical of the polyethylene depend on factors like; “degree and type of branching, crystal structure, and many more. Polyethylene (BE) can be accompanied by or mixed with other materials in many applications to yield improved mechanical and physical properties compared with the base polymer”. Thus, to check the changes in the blended ratios and identify the blend ratio that provides the best mechanical properties for FDM application, different ratios have been used to do needful of finding out the suitable blending technique of PP or SE.

The purpose of this article is to investigate how multi-objective optimization techniques might be applied to machining processes to increase surface quality and productivity. It aims to resolve the competing demands of increasing material removal rates and reducing surface roughness. The paper suggests a hybrid strategy that combines “Genetic Algorithm (GA)” for the improvement and “Response Surface Methodology (RSM)” for process modelling to accomplish this. The methodology aims to quickly and accurately determine the ideal machining parameters. The article structure is as in Section 2, the materials and the experimental setup. In Section 3, the results of the study were discussed, and the conclusion and future aspects of the study will be discussed in Section 4.

2. MATERIALS AND EXPERIMENTAL SETUP

2.1 Materials

The Supplies and equipment that were used in this experiment were polypropylene and sustainable epoxy pellets. These pellets were combined and blended in an extrusion procedure to generate tensile samples. Depicts the entire process of converting waste cooking oil with the condensation method utilizing Bio Monomer with a mix and pour of PP and SE to obtain filament after the extrusion process and then using 3D printing to implement the required application (see Fig 1). Hence, the compositions of PP-polypropylene and SE-sustainable epoxy are shown in Table 1.

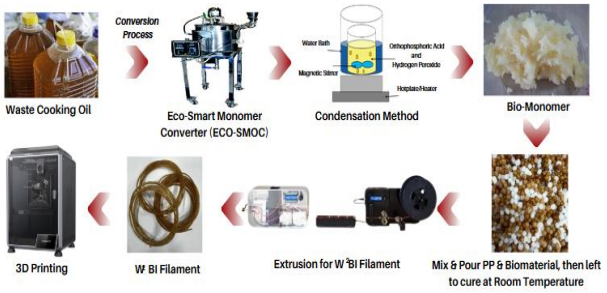


Fig. 1 The Flow process of Converting Waste Cooking Oil with the Condensation Method of 3D Printing.

Table 1. Composition of polypropylene and sustainable epoxy (weight percentage, %).

Sample	PP	SE
PX 0	100%	0%
PX 1	99%	1%
PX 2	98%	2%
PX 3	97%	3%
PX 4	96%	4%
PX 5	95%	5%

2.2 Filament Extrusion for 3D Printing

To prepare the filament with a standard size of about 1.75mm Felfil Filament Maker filament-making machine manufactured by Felfil Evo Extruder of Torino, Italy. However, this device has a single-screw extruder having

four different parts for heating; the critical characteristics of the instrument include the temperature settings of the heating sections, the speed of the screw as well as the fan speed. It cools the extruded filament with four fan units, regulated with a flow deviator to give a uniform cooling in the filament. However, an inhomogeneous cooling would lead to a filament with an oval cross-section due to the maximum level of reduction along the cooling draw direction due to its high crystallinity of PP. The filament is then drawn down with a two-wheel system that rotates in opposite directions, and the speed of these wheels can be regulated either by hand or automatically depending on the required diameter. The filament for PX 0 until PX 5 is shown in Table 2. The parameters used in the extrusion process of the filaments noted above where the results are presented in Table 3 below.

Table 2. Samples of extrusion filament for 3D printing

	Filament			
Samples	PX0		PX3	
	PX1		PX4	
	PX2		PX5	

Table 3. Samples of extrusion filament for 3D printing

Barrel zone temperature (℃)				Screw speed (RPM)	Puller speed (m/s)	Fan speed (%)
T_1	T_1	T_1	T_1			
200	205	210	220	4	0.46	15

2.3 3D Printing by Filament Extrusion (Creality K1, 3D Printer)

In materials extrusion, determining the appropriate printing parameters is important [26]. Initially, the process of polypropylene (PP) extrusion was first optimized [27], following which the parameters and experimental circumstances for the current task were established. Table 3 contains specifics on the conditions that were implemented. When the polymer filaments were placed into the heated printer head, the printing process started. The stepper motor made it easier for the head of the printer assembly to position itself so that it could navigate the molten filament through the liquefier and ensured consistency before it was injected into the nozzle. In order to construct the dumbbells in compliance with ISO 527-2 specifications, the nozzle dispensed the molten PP and SE mixtures onto the printer platform inside the XY plane at an angle of 45 degrees (Table 5) at a specified infilling rate and height along the z-axis. After the entire cross-section was deposited along the XY plane, the print head ascended approximately one layer of thickness (0.20 mm) in the Z dimension. As instructed by the G code (programming language), the entire procedure was carried out repeatedly. Layer-by-layer deposition was the method employed for creating the whole dumbbell (Table 4). The printed dumbbells were used to assess the mechanical properties, microstructure, and morphology of the samples.

Table 4. 3D printing dumbbell samples

	Printed Dumbbell			
Samples	PX0		PX3	
	PX1		PX4	
	PX2		PX5	

Table 5. Printing parameters used to prepare the G-Code for the Creality Slicer Software during 3D printing

Parameters of printing	Designated value
Nozzle temperature	250℃
Print bed temperature	100℃
Diameter of the nozzle	0.4 mm
Thickness of the layer	0.20 mm
Filling percentage	100%
Speed of the printing	30 mm/s

2.4 Accelerated Weathering Test of PX

In the tensile samples, the UV light irradiation was performed using a "UV Accelerated Weatherometer made by Haida International Equipment td. Cabinet number three contained four accelerated weathering tested specimens based on the UV accelerated weathering test conducted as per the ASTM D 4587 which was a standard practice for fluorescent UV-condensation exposure. The UV Weatherometer includes UV fluorescent lamps that produce light of wavelengths of between 280 and 320 nm with a subsidiary at four hundred nanometres. The PSE samples were mounted on a rack holder and UV irradiated at 50℃". One sample of each composition of the dumbbell was subjected to UV weathering after which the changes in mechanical properties of the PX were assessed at 1000, 2000, and 3000 hours.

2.5 Tensile Test

Following irradiation with UV light from 0 to 3000 h "the mechanical properties of the samples of the PX were determined by tensile testing to ISO 527-2. The conditions of the tensile test were a load frame of 5kN

and a cross-head speed of 5.mm/min. The strain was recorded using an extensometer across a gage length of 20 mm". At least five samples per composition were analyzed and an average result was therefore achieved.

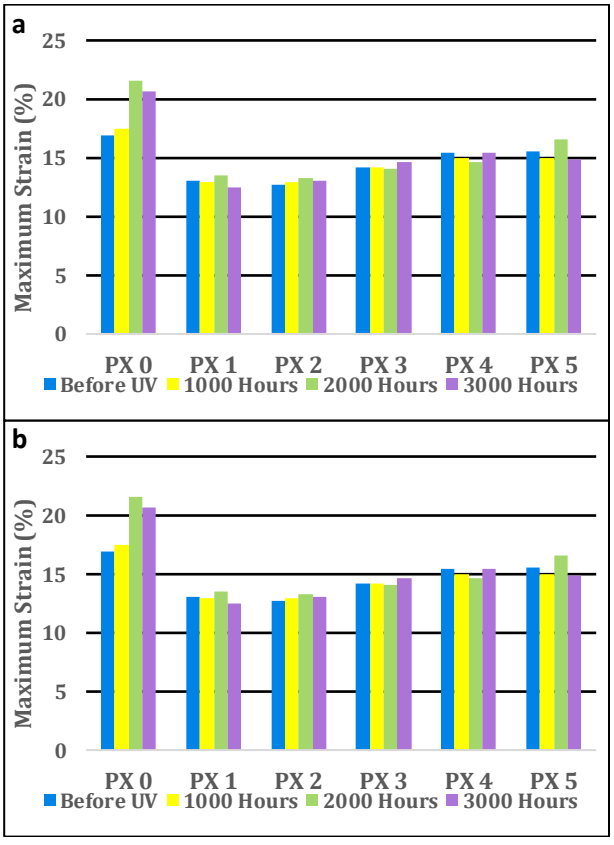
2.6 Scanning Electron Microscope (SEM)

The actual examination of the PX fracture surface is carried out with the aid of a Scanning Electron Microscope Hitachi to carry out the morphological analysis. The microstructure of the fracture surface of PX before and after UV exposure was enlarged to map and compare the weathering impact on the tensile characteristics of PX. The PX thin film samples of the fractured surfaces were initially deposited using an Auto Fine Coater Machine, where the surface of the samples gets coated with a very thin layer of gold at a calibrated current plasma of 25 mA and a chamber pressure of 2 Pa. Images for cellular structures were observed using an SEM of Hitachi at 10 kV and using a 50 µm magnifier under high vacuum.

3. RESULTS AND DISCUSSION

3.1 Tensile Properties

As shown in Appendix 1, the tensile strength of the PX varies from 10 MPa up to 20 MPa. In this present work, tensile strength was found to be the highest (19.11 MPa) for the sample comprising PX 2 with 1000 hours of UV irradiation. This could most probably be due to stiffer polymer grades in the sustainable epoxy resin examined in this study. Consequently, the tensile strength of the PX reduces as the proportion of sustainable epoxy rises. The tensile strength of the sample PX 1 with 1% sustainable epoxy was 18.36MPa to 19.07 MPa and the tensile strength of the sample PX 5 with 5% sustainable epoxy was 14.64MPa to 16.47 MPa. Each PX sample mixture was combined and blended with 1%, 2%, 3%, 4%, and 5% of sustainable epoxy by extrusion and 3D printing.



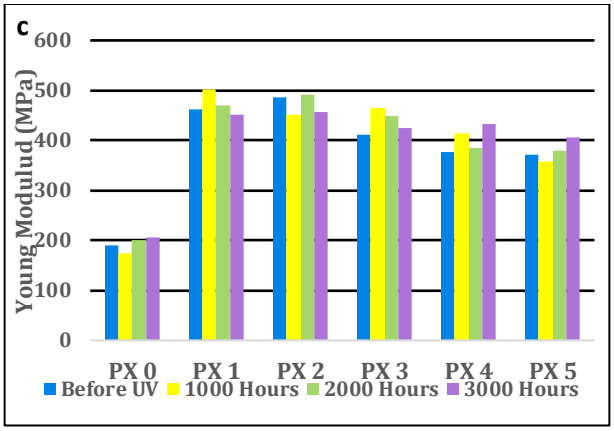


Fig 2. (a)Tensile strength of PX sample before following UV, (b)The highest percentage of PX samples both before and following UV, and (c) The Young Modulus of PX samples before and after UV.

Fig 2(a) displays the strength of tensile of all the PX depicted before and after the UV irradiation test. Polymer composite PX showed better yield stress compared to PX 0 which could be attributed to the combined polymeric type and chemical impurities available in the sustainable epoxy.

As obtained from Fig. 2 (a) and (b), the force of tensile and elasticity upon breaking increases as the percentage of sustainable epoxy increases. This is because sustainability epoxy properties are dominant in influencing the coating resonance. It was also established that an increase in the sustainable epoxy raises the extent of strain and lowers the tensile strength and elongation at break. As it can be observed from the testing results sample PX 5 which contains only 5% of sustainable epoxy has lower strength of the tensile and elongation at break in contrast with sample PX 1 which contains 1% of sustainable epoxy only. On the other hand, Sample PX 0 has a higher strain value than PX due to the absence of sustainable epoxy in its formulation.

This implies that one of how the properties of PX samples can be enhanced is by enhancing the ductility of the matrix material. For every simple composition, a sustainable epoxy of 1%, 2%, 3%, 4% and 5% was added. With the addition of sustainable epoxy to the composition, the polymer composites increased. Tensile strength is depicted in Fig 2 (a), maximum strain in Fig 2 (b), and young Modulus in Fig 4 (c) for the samples UV exposed for 1000, 2000, and 3000 hours. Every graph indicates the deterioration of tensile properties of the sample for PX 1 and PX 2 after UV irradiation because of the discolouration of the composite polymer. As observed, samples having enhanced concentrations of sustainable epoxy, that is PX 3, PX,4 and PX 5, displayed higher inclined tensile performance after the experiment on UV irradiation. For PX 3, the tensile and strain properties were increased while the young modulus was decreased. Besides, PX 4 shows that the tensile properties were decreased, but the strain and young modulus were increased. Sample PX 5 shows an increase in all terms of tensile properties compared to other samples.

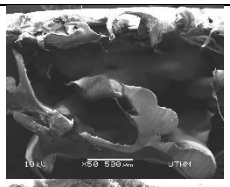
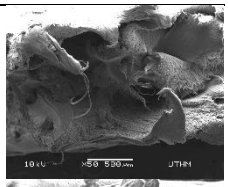
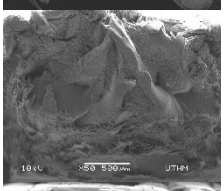
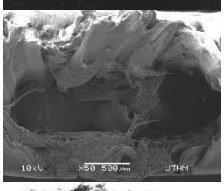
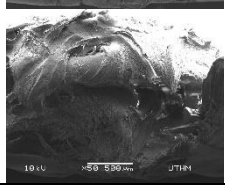
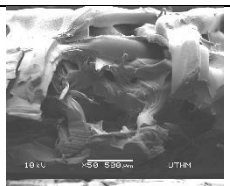
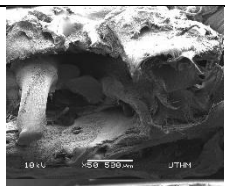
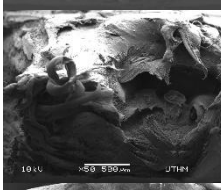
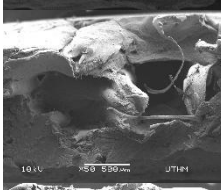
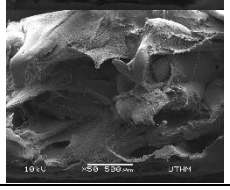
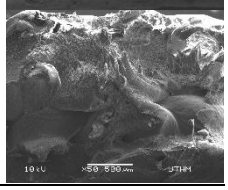
Concerning the percentage of sustainable epoxy (SE), the tensile strength of the epoxy surface rose tremendously as well. According to some data, cellulose and stress transfer efficiency are interrelated variables; thus, when adding SE, crystallinity will be enhanced, and the result will be stiff composites [28]. Thus, the degree of

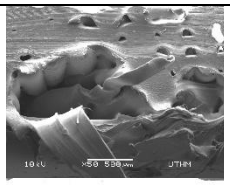
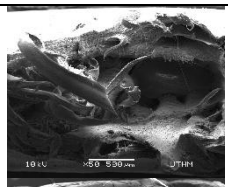
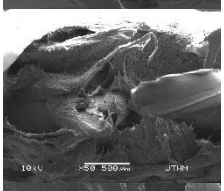
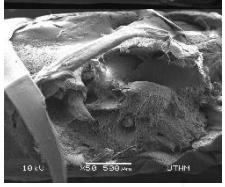
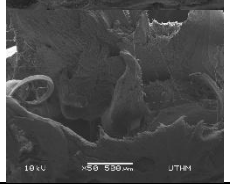
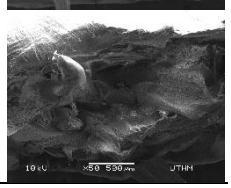
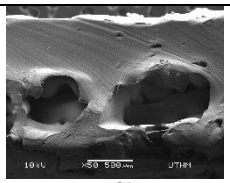
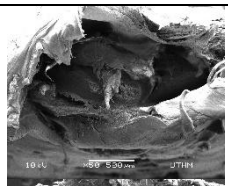
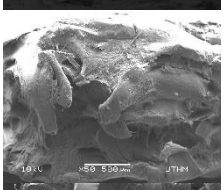
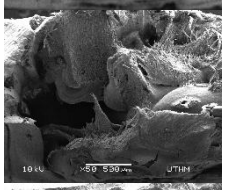
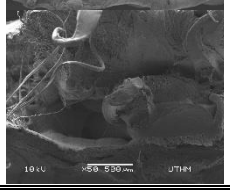
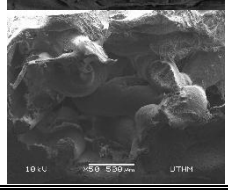
crystallinity of the composites reduced with an increase in lignin loading levels; as evidenced in the tensile strength value of sample PX 5. According to Nikafshar et al. [29], apart from its ability to undergo chemo-crystallization, UV irradiation caused the chain scission of the PP. In addition to that, the weathered cellulose fibre surface has micro-cracks that are not conducive to the transfer of stress from the PP matrix to fibres due to poor tensile strength. Compared with sample PX 3, sample PX 5 which contains a higher content of lignin has relatively high rigidity modulus retention ratios after weathering, possibly because of the cross-linking as well as recrystallization in the PP matrix and the anti-oxidation effect of the lignin [29].

3.2 Characterization of Fracture Surface

The improvement of interfacial bonding in polypropylene (PP) composites by sustainable epoxy (SE) results in improved stress transfer and less void formation, as demonstrated in Table 6, the characterization of fracture surfaces. Therefore, the higher sustainable epoxy content enhances UV resistance and structural integrity but increases cell brittleness due to increased crystallinity.

Table. 6 Demonstrate the crystalline structure of the material's fracture surface following testile testing both before and following UV exposure.

	0 Hours		
PX 0		PX 3	
PX 1		PX 4	
PX 2		PX 5	
	1000 Hours		
PX 0		PX 3	
PX 1		PX 4	
PX 2		PX 5	

	2000 Hours		
PX 0		PX 3	
PX 1		PX 4	
PX 2		PX 5	
	3000 Hours		
PX 0		PX 3	
PX 1		PX 4	
PX 2		PX 5	

In the samples of the present work, where PX is equal to zero, the character of fractography of the surface is relatively homogeneous and the matrix has been pulled out. The zones of voids and fractures are more numerous in the tensile separation area of the sample PX 0 than in the tensile separation area of the sample with 1% SE (PX 1) having fewer voids and fibre breakage. This is well illustrated when SE rises from 1% to 2%, in which the void cavity and fibre breakage are relatively rare. As the percentage of SE increases, admissible and actual interaction between the filler and the matrix polymer becomes negligible. Of these, the segregated and separate spaces observed between, the matrix and the fibres require a wrong bond. For samples, PX 3, PX 4, and PX 5, the crack passing through the sample shows stress transfer from matrix material to the SE Fiber. An increase in the interfacial adhesion between the filler and the PP matrix due to the esterification mechanism results in a matrix crack at the filler itself [30]. In other words, stress is transferred well from the filler to the matrix polymer, which as a result enhances the modulus of the stress on it. Sample PX 5 possesses a very low degree of torn matrix across the entire fracture surface. This means that the sample which has a greater percentage of rice husk content is much more brittle.

SE was included in every single one of the compositions starting from PX 1 until PX 5. Each composition was mixed and blended with 1%, 2%, 3%, 4%, and 5% of SE. Subsequently, the increase in the percentage of SE enhanced the bonding between SE and PP matrix to a reasonable extent. Observations made from the SEM images reveal that with the increase in the percentage of PP in the SE composites, the separation between the wood flour and the polymer matrix is difficult to see, which suggests a better interface adhesion.

The impact of weathering on the mechanical properties and morphology of the fracture surface was studied by weathering each sample at 1000, 2000, and 3000 hours using a UV irradiator. Subsequently, all the composites underwent degradation after being subjected to the process of weathering. Even though several matrixes are pulled out in the fracture surface of sample PX 1; the cavities are only slightly visible after being exposed to 3000 hrs of UV irradiation. As there is 5% SE in sample PX 5, there are little fibres pull-out on the fracture surface. However, in the concerning samples PX 1 and PX 2, the pull-out was found to influence the large fibres only, where SE % is respectively 1% and 2%.

4. CONCLUSION AND FUTURE TRENDS

The study's key involvement is the establishment and use of a novel hybrid optimization framework that combines GA with RSM. This integration makes it possible to solve the multi-objective optimization issue efficiently and simulate the machining process precisely. Additionally, by balancing competing, the objective optimization approach provides insightful information for industrial applications. In this study, we employed a novel strategy which by first converting waste cooking oil (WCO) and then combining it with photopolymerizable monomer to produce SE-monomer, which is an instance of mechanically enhanced and cost-effective SE to 3D printable photocurable resin for 3D printing. The results of the experiments showed that an appropriate SE can be permanently present in 3D printed products, significantly increasing the mechanical strength of the PX polymer composite system while effectively resolving the low toughness and high brittleness that are inherent in PX polymer composite systems. In this system, SE filler functioned comparably to a PX polymer composite.

Moreover, given that WCO is utilized as a raw material, adopting reasonably priced SE as a filler decreases the material's overall manufacturing cost relative to other materials, which strengthens the material's value and competitiveness in the market. Consequently, the conversion of WCO into SE filler 3D printing has facilitated the high-value utilization of SE and has considerable potential for facilitating the development of affordable, environmentally conscious, and sustainable 3D.

Future Trends within 3D printing include enhancing interlayer adhesion and mechanical qualities, investigating multi-material printing processes, and emphasizing recycling and circular economy ideas. Green manufacturing techniques such as 3D printing using renewable energy are being developed to grow sustainability and commercialize sustainable composites. This involves collaborations between scientists, engineers, and industry competitors.

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