Research Article

Recycling of Polycarbonate/Acrylonitrile Butadiene Styrene Blends with Flame Retardant Additives for 3D Printing Filament

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Received 6 December 2023; Revised 14 March 2024; Accepted 22 March 2024; Published 28 March 2024

Academic Editor: Sohaib Z. Khan

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Recycling waste from electrical and electronic equipment (WEEE) for valuable materials is a critical aspect of the circular economy and sustainable waste management. Recycling these solid wastes into value-added products is one method for reducing landfill waste and pollution. The purpose of this research was to investigate the recyclability of plastic wastes of Polycarbonate Acrylonitrile Butadiene Styrene blends with Brominated Flame-Retardant additives (PC-ABS-FR). Two types of plastic wastes, PC-ABS-FR (GL) and PC-ABS-FR (DB), were collected and converted into polymer chips using a mechanical process. To study the effectiveness of waste recyclability, 3D printing filaments were produced through the extrusion process. Chemical elements, functional groups, and thermal and mechanical properties of recycled products were evaluated using EDX, FTIR, DSC, and universal tensile testing equipment, respectively. TGA, cone calorimetry, and melt flow index analyses were also carried out. The results showed that recycled PC-ABS-FR (GL) waste-based filament samples had better thermal and mechanical properties. On the contrary, PC-ABS-FR (DB) waste recyclability was not effective in filament extrusion. The mechanical properties of the 3D printing filaments produced from PC-ABS-FR electronic waste were found to be similar to those observed in commercially available and previously reported filaments. The strength, thermal property, cross-sectional homogeneity, and filament diameter of 1.8 ± 0.03 mm of the 3D printer filament made from PC-ABS-FR (GL) were suitable for 3D printing applications such as children’s toys, dust bins, and flower vases.

1. Introduction

Recycling of plastic waste plays a significant role in circular economy and sustainability practices [1]. Recycling plastic is helpful for resource efficiency by lowering the need for new raw materials, thereby addressing resource scarcity and promoting sustainability [2, 3]. Plastic recycling can also help mitigate environmental pollution and improve waste management processes [4]. Recycled plastics are valuable materials covering a wide range of applications in everyday life and can be found everywhere, from households to industry. Vital economic activities as diverse as packaging, construction, transportation, healthcare, and electronics depend heavily on plastics as raw materials [5]. Due to the considerable increase of waste from electrical and electronic equipment (WEEE) in the world each year, further efforts have been made to recycle materials. The management of electronic waste (e-waste) is a significant topic for consideration on the circular economy agenda [6]. Studies carried out in 2016 show that on a global scale, 15 million tons of waste from the total WEEE was occupied by plastic materials [7, 8]. Disposable plastic materials are commonly used in printer machines, and the generation of solid waste is substantial due to their short life cycle [9, 10]. Of the total WEEE, 25% is occupied by plastic materials, from which a significant share was taken by waste PC-ABS blends [11]. To overcome the problem, recycling waste plastic materials was taken as the best option. Several advantages will be obtained through the recycling of materials [12]. Recycling metals from WEEE requires significantly less energy compared to virgin metal production, leading to lower greenhouse gas emissions and reduced pressure on the energy grid.
The constant evolution of electronic devices necessitates continuous adaptation of recycling technologies to handle new materials and components [14]. The development and marketing of recycled plastic extruders, which produce 3-D printing filament from recycled materials, have significantly broadened the range of materials available for consumer 3-D printers [15]. Additionally, WEEE also holds expensive materials, including rare earth metals, and recycling offers the possibility of reusing these materials again. Plastic recycling can be done either energetically, chemically, or mechanically [16].

Nemours researchers developed 3D printing filament from recycled plastics such as high-density polyethylene [17], polypropylene [18], and polyethylene terephthalate [19]. The most used materials in fused deposition modeling (FDM) are Acrylonitrile Butadiene Styrene (ABS), polylactic acid (PLA), and polycarbonate (PC) [20–22], ABS/Silicon [18]. Dani et al. conducted a study on the ABS polymeric material derived from the computer cover and examined its recycling process under various processing conditions. The findings suggest that a significant decrease in impact strength was observed, likely attributable to the molecular crosslinking and chain fragmentation within the rubber component [23]. Imbernon et al. investigated on processing of flame-retardant additive-free pellet PC-ABS using an injection molding process. The findings showed that the tensile properties of products were significantly affected by the product cycle [24]. Woern et al. discovered that recycled waste plastic has the potential to be transformed into filament for 2.5 cents per kilogram, making it significantly less expensive compared to the commercial filament. This process involves producing filament from polymers with extrusion temperatures below 250°C, allowing for the creation of customized filament using various homopolymers and composites. This development has significant implications for material science research, recyclability studies, and exploring new applications of fused filament-based 3-D printing [25].

Chiu et al. investigated the characterization of a PC/ABS blend over 20 reprocessing cycles and the subsequent restoration of its functionality using virgin additives. The recovery of the waste material, which had been reprocessed 20 times, was accomplished by introducing approximately 30% (by weight) of virgin PC and ABS, along with 1.5% (by weight) of a chain extender and 2% (by weight) of styrene maleic anhydride, simultaneously [26]. Cafero et al. analyzed WEEE plastics and their potential for 3D printing filaments. They found that plastic is the most challenging fraction of WEEE to manage, and the market for recycled plastics from WEEE is limited. WEEE samples showed good similarity to virgin polymers with fewer halogens and inorganic fillers. Test objects printed with WEEE filaments showed no significant deviation from model design compared to commercial filaments [27]. Chawla et al. fabricated fused filaments for the FDM technique of additive manufacturing from recycled ABS by using a twin-screw extruder. Thermal analysis performed to estimate the heat-carrying capacity of recycled ABS highlighted that the heat capacity of ABS increases significantly from 0.28 J/g to 3.94 J/g during the heating cycle [28]. Oliveira et al. tested how recycled ABS (rABS) from e-waste was mixed with virgin ABS (vABS) at various ratios for 3D printing filament application. Differential scanning calorimetry revealed that the glass transition temperatures for vABS/ rABS blends were between those of vABS and rABS. Torque rheometry investigation revealed that the addition of rABS did not affect the processability of vABS. Increased rABS content up to 50 wt% resulted in higher impact strength (IS) in 3D printed samples [29]. Mishra et al. produced and analyzed 3D printing filaments made from recycled/virgin ABS mixes, and the results revealed that the breakdown of the styrene-acrylonitrile and butadiene components in ABS induces strain hardening and material stiffening. The 80% rABS/20% vABS filament has a Young’s modulus of 2329 MPa, yield strength of 34.814 MPa, and ultimate tensile strength of 40.82 MPa, similar to vABS filament [30].

1.1. Research Objective. As technology advances, the volume of electronic plastic waste, such as PC-ABS waste, is increasing. However, the correct recycling methods and applications for this waste are still a matter of concern. In particular, the mechanical recycling of waste PC-ABS mixes with flame-retardant additives for 3D printing applications has not yet been thoroughly investigated. Therefore, the primary focus of this research is to study the mechanical recycling of waste polycarbonate/acrylonitrile butadiene styrene blends with brominated flame-retardant additives (PC-ABS-FR), a topic of significant importance in the field of plastic waste recycling.

2. Materials and Methods

2.1. Materials. A scrap of waste PC- ABS–FR with product code RC3-0794 and material code > PC + ABS-FR (40) <=1 or in short symbol PC–ABS–FR (GL) Moreover, dark black color waste PC–ABS–FR with product code RC3-0792 and material code > PC + ABS-FR (40) <=1 or explained in short form as PC–ABS–FR (DB) materials were selectively taken from HP printer hardware cover from waste printer store.

2.2. Waste Preparation and Recycling. The procedure that was utilized for the characterization of PC–ABS–FR consists of sorting of blends, cleaning or washing of sorted material for removal of organic and inorganic contaminants, size reduction (flake production), plastic processing (remelting), and sample preparation for characterization. Material identification was done by looking at the tag/trademark stamped on the surface of the sample, and further approval was performed using FTIR analysis. Following the identification of the materials, cleaning was carried out using 300 g detergents to remove the surface impurities of the samples. Finally, washed samples were placed in an oven at 50°C temperature for drying and, successively, to prepare for size reduction, obtaining approximately 7–10 mm flakes in order to continue analysis (see Figure 1).
2.3. Extrusion of Recycled Granules and Filament Manufacturing. Thermo-Hake PTW16 intermeshing corotating twin screw extruder with a screw diameter of 16 mm and die diameter of 3 mm was used for the extrusion of flakes of waste PC-ABS-FR samples (Figure 2(a)). The processing temperature was regulated in order to obtain a final diameter of the extruded filament of $1.75 \pm 0.10$ mm targeted for 3D printing application. Extrusion analysis of PC-ABS-FR (GL) and PC-ABS-FR (DB) was carried out. However, because of poor results obtained through many trials, only PC-ABS-FR (GL) extruded filament products were reported in Figure 2(b).

2.4. Extruded Filament Analysis. The chemical composition, functional groups, and thermal and mechanical properties of recycled plastic materials were assessed using EDX, FTIR, TGA, DSC, and tensile strength tests. These evaluations play a critical role in determining the quality and characteristics of recycled plastic products [31, 32].

2.4.1. FTIR Analysis. FTIR analysis was measured using Perkin Elmer ATR-FTIR Spectrum diamond-covered reflection to characterize functional groups of the PC-ABS blend. Here, the identification of samples was done by directing infrared rays toward the sample and looking at the vibration of their chemical compounds. The scan was done between 4000 cm$^{-1}$ and 600 cm$^{-1}$ averaged for each spectrum at intervals of 1 cm$^{-1}$ with a resolution of 4 cm$^{-1}$.

2.4.2. EDX (Energy Dispersive X-Ray Analysis). Analysis of the chemical composition of waste PC-ABS-FR plastics was carried out by using an SEM Jeol IT300 instrument. It is done to detect compositions of toxic and environmentally hazardous elements that may originate from additives added during the manufacturing of products.

2.4.3. TGA (Thermo-Gravimetric Analysis). Weight loss as a function of the temperature of waste PC-ABS-FR plastics was analyzed using a Q5000 IR thermos-gravimetric analyzer. Sample weight of 10 mg waste PC-ABS-FR was taken, and analysis was done with an airflow rate of 15 ml/min, and temperature ranging from RT to 700°C at the rate of 10°C/ min was used during analysis. Intersections of the two tangent lines were taken, and the onset of degradation temperature was evaluated.

2.4.4. DSC (Differential Scanning Calorimetry). Heat flow into and out of the system as a function of the temperature of samples was evaluated using a Mettler DSC 30 Swiss Mettler Toledo calorimeter. During the test, 10 mg of samples of plastic were taken, and a nitrogen flow of 100 ml/min was applied. Meanwhile, the glass transition temperature (Tg) of waste PC-ABS-FR samples was measured by taking thermogram inflection points.

2.4.5. Cone Calorimetry Analysis. Cone calorimeter analyses of waste PC-ABS-FR plastics were performed to verify the existence of flame-retardant additives within the samples. Here, material flammability parameters, namely, time to ignition (TTI), heat release rate (HRR), peak heat release rate (PHRR), total heat release (THR), and final mass of samples, were analyzed beyond verification of flame-retardant additives.

2.4.6. Melt Flow Index Analysis. Melt flow index analysis (MFI) of PC-ABS-FR plastics was done using Kayeness Capillary Rheometer (Model 4003DE, Morgantown, PA, USA) according to the ASTM D1238 standard. During measurement, the barrel length of 162 mm, barrel diameter of 9.55 mm, die length of 8.0 mm, die diameter of 2.096 mm, load of 5 kg, sample weight of 5 g, and melting temperature of 200°C were used for analysis of both PC-ABS-FR items. Five test replicates were taken in each type of plastic.

2.4.7. Mechanical Properties. Tensile strength and modulus of produced 3D printer filaments from recycled PC-ABS-FR plastics were done using an Instron 5969 universal tensile strength machine with a 50 KN load cell with an extension speed of 0.25 mm/min as instructed in the ASTM D638 test method.

3. Results and Discussion

3.1. FTIR Analysis Result. For comparison purposes, two additional samples, namely, waste PC-ABS and Pellet PC-ABS-FR, were analyzed together with PC-ABS-FR (GL)
and PC-ABS-FR (DB). The spectrum peaks of both PC-ABS-FR (GL) and PC-ABS-FR (DB) were reported in Figure 3. The presence of ABS was seen from the characteristic vibrations between 2967 and 2853 cm\(^{-1}\) (C-H stretch vibration), 1592–1504 cm\(^{-1}\) (C=C aromatic stretch vibration), at 1409 cm\(^{-1}\) (C-H\(_2\) and CH\(_3\) bending stretch vibration), 960–950 cm\(^{-1}\) (out of phase or C-H bending vibration tetra substituted C=C, Trans structure). In addition, 760 and 698 cm\(^{-1}\), peaks are expected to be chloride vibration peaks that may originate from flame-resistant additives. On the other hand, the presence of PC was seen by looking at the characteristic vibration of ester bends between 1766 and 1769 cm\(^{-1}\) (C=O stretch vibration), at 1592 cm\(^{-1}\) (C=C stretch vibration of aromatic ring structure), 1080–1013 cm\(^{-1}\), and 1187–1158 cm\(^{-1}\) (C-O stretch vibration of ether/ester functional groups).

3.2. EDX Analysis Results. After taking 20 mg waste PC-ABS-FR samples and performing EDX analysis, elemental compositions were presented. As shown in Figure 4 and Table 1, waste PC-ABS-FR (GL) contains more bromine, i.e., 0.96%, in comparison to PC-ABS-FR (DB), which is 0.07%. However, PC-ABS-FR (DB) contains more titanium (0.06%) and phosphorus (0.49%) compared to PC-ABS-FR (GL), according to the European directive (12). The threshold value for toxic elements, particularly Cd, Br, Cr, Ti, P, and Hg, must be 0.1% for recycling. The chemical compositions of toxic elements in samples are below the minimum threshold level, and the recyclability of waste samples is not affected [33].

3.3. TGA Analysis Result. From the thermogram, taking the intersection of the two tangent lines, the onset of decomposition of the two samples, namely, PC–ABS–FR (DB) and PC–ABS–FR (GL), was analyzed, and the final result is indicated in Figure 5. Two-stage thermal degradation peaks have been seen in both PC–ABS–FR (DB) and PC–ABS–FR (GL). In the case of PC–ABS–FR (DB), the first degradation starts at 200°C and ends at 473°C. However, the second 2nd stage degradation starts from 475°C and ends at 550°C. On the other hand, in the case of PC–ABS–FR (GL), the first phase degradation starts at 225°C and ends at 425.5°C. However, the 2nd stage degradation takes place between 425.5°C and 550°C. In both cases, the first phase degradation corresponds to the degradation of ABS, and the 2nd phase degradation corresponds to the degradation of the PC component [34–36]. Further decomposition has been seen in both samples, starting from 550°C to 675°C. This further decomposition may have been expected to be the decomposition of additives added during plastic manufacturing. The onset degradation temperature of both wastes of PC-ABS blends is around 50% lower than the pellet one, and it is expected to be the effect of aging of materials. Since the melting temperatures of ABS and PC plastics are approximately 200°C and 475°C, respectively [37,38], from a thermal resistance point of view, recycling of waste PC-ABS-FR(GL) sample is comparatively more acceptable.

3.4. DSC Analysis Result. Figure 6 shows the DSC analysis of PC-ABS-FR (GL) and PC-ABS-FR (PL) samples of thermograms. The thermal transition results of the two samples are reported in Table 2. From the first heating thermograms of PC–ABS–FR (GL) and PC–ABS–FR (PL), two peaks were differentiated from each item, i.e., at 104°C and 151°C from PC–ABS–FR (GL) and at 114°C and 158°C from
PC–ABS–FR (PL) which corresponds to the glass transition temperature of ABS and PC phases, respectively, and the result was noted by taking the midpoint of infection. However, in the second scan, heating thermal transitions were seen between 95°C and 112°C in PC–ABS–FR (GL) and PC–ABS–FR (PL), respectively. This transition is expected to be the glass transition of the ABS phase of samples remaining from the first scan. Here, the onset of the glass transition temperature of ABS in the second scan is 5–10% lower than the pellet one, which is 100–105°C [39]. A designer has to take this reduction into account during product design. On the other hand, the glass transition temperature was seen during the cooling scan, and it is expected to be the glass transition of additives added during product manufacturing. Finally, the glass transition temperature of PC–ABS–FR (GL) is nearly consistent with the glass transition temperature of version (pellet) samples; recycling is expected to be acceptable.

3.5. Cone Calorimetry Analysis. Cone calorimeter analysis results of PC-ABS-FR (GL) and PC-ABS-FR (DB) were reported in Table 3. The cone calorimeter result confirms that the two waste PC-ABS blends consist of brominated flame retardants with Sb₂O₃ + TiO₂.

3.6. Melt Flow Analysis. Table 4 shows the melt flow of control and recycled plastics (PC–ABS–FR (DB) and PC–ABS–FR (GL)). The test was carried out according to the ASTM D1238-A standard using a melt flow rate in g/10 min, temperature of 260°C, and load of 2.16 kg for three samples, namely, PC-ABS-FR (DB), PC-ABS-FR (GL), and PC-ABS-FR (PL). The melt flow rate of waste PC-ABS-FR (GL) and PC-ABS-FR (DB) samples shows approximately 3.5% higher melt flow rate in comparison with PC-ABS-FR (PL). Since the viscosity of amorphous polymers was primarily influenced by the age of the materials (20), the slight increase
Figure 5: (a) Thermogravimetric (TG) and (b) (DB) differential thermal gravimetric (DTG) curves.

Figure 6: Thermo-grams of PC-ABS–FR (GL) and PC-ABS–FR (PL) samples.

Table 2: Thermal transition of PC-ABS-FR granule powder.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Tg of 1st scan heating (°C)</th>
<th>Tg of 2nd scan heating (°C)</th>
<th>Cooling</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Tg₁</td>
<td>Tg₂</td>
<td>Tg₁</td>
</tr>
<tr>
<td>PC-ABS-FR (GL)</td>
<td>104.2</td>
<td>151.3</td>
<td>95.1</td>
</tr>
<tr>
<td>PC-ABS-FR (PL)</td>
<td>114.3</td>
<td>158.2</td>
<td>112.5</td>
</tr>
</tbody>
</table>

Table 3: Cone calorimetry analysis results of PC–ABS samples.

<table>
<thead>
<tr>
<th>Samples</th>
<th>Fluorescence X</th>
<th>Cone</th>
</tr>
</thead>
<tbody>
<tr>
<td>PC–ABS–FR (GL)</td>
<td>Brominated FR with Sb₂O₃ + TiO₂</td>
<td>Confirmation of FR</td>
</tr>
<tr>
<td>PC–ABS–FR (DB)</td>
<td>Brominated FR with Sb₂O₃ + TiO₂</td>
<td>Confirmation of FR</td>
</tr>
</tbody>
</table>
in the melt flow rate of waste PC-ABS-FR samples mainly corresponds to the aging effect of materials. This increase in the melt flow rate can be adjusted by controlling process parameters such as temperature and operating time.

Table 5 shows the linear density, diameter, and productivity of 3D printer filament made from PC–ABS–FR (GL). Similar temperature settings were used during the extrusion of both PC–ABS–FR (DB) and PC–ABS–FR (GL), and the setting was adjusted according to the extrusion temperature setting for PC–ABS blends [40]. Here, the initial temperature setup ($T_1$) was modified by looking at results obtained after the first and second trials. A gradual increase in melting temperature was carried out in order to get filaments of uniform and suitable diameter for 3D printing applications [41]. Finally, the PC–ABS–FR (DB) samples do not have a regular cross section, and there is no continuity in filaments. However, PC–ABS–FR (GL) shows the best cross-sectional homogeneity and continuity during filament production.

### Table 4: Melt flow analysis results of PC-ABS-FR samples.

<table>
<thead>
<tr>
<th>Samples</th>
<th>Melt flow rate (g/10 min), 260°C, 2.16 kg</th>
</tr>
</thead>
<tbody>
<tr>
<td>PC–ABS–FR (GL)</td>
<td>22 ± 0.15</td>
</tr>
<tr>
<td>PC–ABS–FR (DB)</td>
<td>21 ± 0.14</td>
</tr>
<tr>
<td>PC–ABS–FR (PL)</td>
<td>16 ± 0.13</td>
</tr>
</tbody>
</table>

### Table 5: Extrusion analysis results of PC-ABS-FR (GL) sample.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Temperature setting zones (°C)</th>
<th>Linear density (g/m)</th>
<th>Productivity (g/min)</th>
<th>Average diameter (mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PC-ABS–FR (GL)</td>
<td>130 200 200 205 200</td>
<td>0.33</td>
<td>9.83</td>
<td>1.8 ± 0.03</td>
</tr>
</tbody>
</table>

### Table 6: Tensile strength and extension of recycled PC-ABS-FR filaments.

<table>
<thead>
<tr>
<th>Sample</th>
<th>$\sigma_b$ (Mpa)</th>
<th>$\epsilon_b$ (mm/mm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PC-ABS–FR (GL)</td>
<td>52.00 ± 1.70</td>
<td>0.03 ± 0.01</td>
</tr>
<tr>
<td>PC-ABS–FR (DB)</td>
<td>34 ± 0.12</td>
<td>0.1 ± 0.01</td>
</tr>
</tbody>
</table>

3.7. Mechanical Properties of 3D Printer Filaments. Tensile strength and modulus analyses of recycled compression molded PC–ABS–FR samples were carried out by using an Instron 5969 electromechanical testing machine with a 50 KN load cell and stress-strain curve, which is reported in Figure 7 and Table 6.

Young’s modulus of tensile dumbbell-shaped plastic products was carried out by taking the first linear segment (slope) of the stress-strain curve through a linear fitting.
As a result, as indicated in the result section, the tensile strength of PC–ABS–FR (GL) is comparatively better than PC–ABS–FR (DB). Since the tensile strength and modulus of PC–ABS primarily depend on the ratio of mix of PC to ABS and the anisotropic nature of the product, this difference in tensile strength and modulus of products were expected to be primarily anisotropy and mix ratio beyond effects of processing machine, molding process, orientation, and properties of fed materials. Table 7 presents the statistical analysis of the mechanical properties of recycled 3D printing filaments. The result of the analysis of variance (ANOVA) indicates that the tensile modulus, tensile strength, and extension of the two types of samples displayed statistically significant differences, with $P$ values of 0.002, 0.003, and 0.001, respectively, at a significance level of 0.05.

The 3D printing filaments that were produced were compared to those available commercially and those previously published. The results are presented in Figure 8. It was found that the filament made from recycled PC-ABS-FR (GL) exhibited a higher strength of 52 MPa, which is comparable to most reported results except for PLA, which had a strength of 65 MPa. Additionally, the tensile strength of the PC-ABS-FR (DB) sample was better than the reported results of PP, HIPS (Blue), and Nylon-618 filaments. In conclusion, the 3D printing filaments made from PC-ABS-FR electronic waste demonstrated mechanical properties that were comparable to both virgin and recycled filaments.

### 4. Conclusions

The main aim of this work was to investigate the mechanical recycling of two specific waste PC–ABS blends, namely, PC–ABS–FR (DB) and PC–ABS–FR (GL), through chemical (FTIR, EDX), thermal (TGA and extrusion) and mechanical such as tensile strength and modulus analysis. In this study, the recyclability of the two waste samples was checked by using the same process parameters and machine. The waste PC-ABS blends are thermoplastic polymers, and their applications were always designed far below the glass transition temperature; the results of TGA and DSC analysis of the two samples were acceptable for recycling waste materials. However, during extrusion, waste PC-ABS-FR (DB) shows high diametrical homogeneity and melts flow ability. In this regard, the material is not suitable for filament production. On the other hand, PC–ABS–FR (GL) shows a prominent result during extrusion, which is $1.8 \pm 0.03$ mm; the product was suitable for 3D printing of products, particularly children’s toy cars. In general, light gray color waste PC–ABS–FR, having product code RC3-0794 and material code PC+ABS-FR(40) or symbolized as PC–ABS–FR (GL) in this paper, are promising materials with better melt flow ability and cross-sectional uniformity for 3D printing of a variety of products.

### Data Availability

The data are available upon request to the authors.

### Conflicts of Interest

The authors declare that they have no conflicts of interest regarding the publication of this paper.

### Acknowledgments

The project was supported by the Ethiopian Institute of Textile and Fashion Technology, Bahir Dar University.

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