Recycling Polymer Blend made from Post-used Styrofoam and Polyethylene for Fuse Deposition Modelling

E P Yap¹, S C Koay¹*, M Y Chan², H L Choo¹, T K Ong³, K Y Tshai¹

¹ School of Computer Science and Engineering, Faculty of Innovation and Technology, Taylor’s University Lakeside Campus, No. 1, Jalan Taylor’s, 47500 Subang Jaya, Selangor, Malaysia.
² Faculty of Engineering and Technology, Tunku Abdul Rahman College University, 53300 Kuala Lumpur, Malaysia.
³ Department of Mechanical, Materials and Manufacturing Engineering, The University of Nottingham Malaysia Campus, Jalan Broga, 43500 Semenyih, Selangor Darul Ehsan, Malaysia.

* E-mail: seongchun.koay@taylors.edu.my

Abstract. Styrofoam is widely used as packaging material for many applications like home furniture and electrical appliance. Styrofoam is a non-biodegradable material which its disposal causes serious environment issues. This research demonstrates an alternate recycling method of Styrofoam waste by converting it into 3D printing filament for Fused Deposition Modelling (FDM). For this research, the recycled polystyrene (rPS) was extracted from Styrofoam waste and blended with low-density polyethylene (LDPE), then extruded into filament using a filament extruder. The formulated rPS/LDPE blend with different blend ratio exhibited a good printability when the printing temperature and extrusion rate fixed at 240°C and 120%. However, the tensile strength of printed specimens with rPS/LDPE blends were lower than printed specimen with neat rPS. The tensile strength and modulus of printed specimens with rPS/LDPE were decreased due to the increase of LDPE content. The decrease of tensile strength mainly caused by the incompatibility between the rPS and LDPE phases. However, the addition of more LDPE content in the blend enhanced the ductility of rPS/LDPE blends. Furthermore, the increase of LDPE content also increased the thermal stability of rPS/LDPE blends. Overall, the rPS/LDPE blend is a potential alternate material for producing FDM filament.

1. Introduction

3D Printing is often defined as a process that involves in the construction of 3D objects with material depositing layer by layer using machines that controlled by computer. The examples of 3D printing, such as Fused Deposition Modelling (FDM), Stereolithography (SLA) and Digital Light Processing (DLP) are widely used in many industries. The 3D printing technology is getting popular among industries as the users can build components due to several benefits, such as mass customization, freedom in design, even if it is geometrically complex [1]. By comparing 3D printing to the other established traditional manufacturing techniques like CNC, injection moulding or casting using mold, the 3D printing technique offers shorter fabrication time, less investment cost and minimize material wastage [2]. Due to many advantages offered by the 3D printing technology, now many industries adopted 3D printing technology in this manufacturing process. As reported by Bankole et al. [3], the
3D printing industries and technologies communities play a role in producing medical and protective devices to deal with the recent Covid-19 pandemic. For instance, Additive Manufacturing Solutions (PTY) Limited in South Africa mass produces face shield frames for medical practitioners using the 3D machine.

Fused Deposition Modelling (FDM) is one of the most widely used 3D printing methods for end users [4]. FDM is famous among many industries because of easy handling, low cost, and the printed part has high geometric accuracy. The FDM printer uses coiled filament made of thermoplastic that is fed through an extrusion nozzle. The filament will be melted at nozzle and extruded to the base platform. The translation pathway of the nozzle will be led by computer-aided design (CAD) program following the X, Y and Z coordinates that designed to build the 3D object. The relatively low cost of its material and simple operation and maintenance of the FDM made it one of the most widely used 3D Printing methods [1]. Due to the flexibility and capability of FDM technology to produce complex geometries, the FDM can produce customized products for various applications such as beauty products, automobile parts, interior design components as well as medical implants [3]. For example, Philips company fabricated customized pendants and table lamps cover for customers using FDM machine.

A wide range of thermoplastics materials that can be printed using FDM machine included general thermoplastic (e.g., ABS, PLA) and engineering thermoplastics (e.g., Nylon, TPU and PETG) as well as high performance thermoplastics (e.g., PEEK and PEI) [1]. However, most of the thermoplastic materials are mainly produced from fossil fuels. The fossil fuels are limited resources in the world, and it might possibly deplete in the future. Moreover, massive amount of plastic waste is threatening the world as the poor waste management methods and low recycling rate of plastic, thus it always causing serious environmental issues. The production of FDM filament from recycling thermoplastics is one of the potential solutions to be an alternative source of material for FDM feedstock, thus the source of FDM feedstock not highly depending on fossil fuels [5-6]. Besides, the utilizing of recycling thermoplastic as feedstock for FDM also promotes the recycling of plastics and it may further reduce the environment impact from plastic wastes.

Polystyrene (PS) foam is known as Expanded Polystyrene (EPS). The EPS material is often used as the protective packaging material on products like fruit, vegetables, electrical appliances, and furniture [7]. The production of EPS increased drastically in the past two decades due to its wide range of applications. For this reason, the accumulated amount of post-used EPS was escalated alarmingly as its almost nonexistence of biodegradation [8]. Feasible measures like avoiding unnecessary uses of EPS and recycling EPS are essential to deal with the environmental issues caused by waste EPS. However, the recycling of EPS is uncommon as compared to the recycling of other thermoplastics. In fact, EPS is a light material that contains approximately 95% to 98% of air in the structure [9]. Due to its bulky structure and occupied large volume of space with minimum mass, the transportation of EPS wastes to recycling center would be costly. As the profit of recycling is calculated based on the mass of material, the recycling of EPS waste becomes less attractive [10]. In order to find a new opportunity to promote the recycling of EPS waste, the present project aims to produce FDM filament using EPS waste. Regarding to the research reported by Ng et al. [11], the FDM filament extruded using recycled PS (rPS) obtained from EPS waste was not as desirable as compared to the commercial high impact PS FDM filament. They found the rPS filament was brittle and cause filament breakage during printing process. Hence, this project targeted to produce FDM filament by blending the rPS from EPS waste and low-density polyethylene (LDPE) resin.

As the recycling of post-used EPS is not practiced as much as other thermoplastics, this research was motivated to create FDM filament from rPS obtained from post-used EPS waste. This project was studied the printability of the rPS/LDPE blend filament with different blend ratios and different 3D printing parameters, such as printing temperature and extrusion rate. The rPS/LDPE blend filament produced in this research also 3D printed specimens and tested for the tensile properties. The thermal stability of rPS/LDPE blend filaments with different blend ratios were also examined in this study.
2. Methodology

2.1. Preparation of Raw Material
EPS wastes were obtained from electrical appliance shops nearby Subang Jaya area. Then, the collected EPS wastes were cleaned using clean wet cloth and left to air-dry under room temperature. Then, EPS wastes were broken into smaller pieces with size smaller than 20 cm$^3$ by hand. The EPS pieces were shrunk into rPS crumbs in the oven at 140℃ for 15 minutes. After cooling down to room temperature, the rPS crumbs were ground into rPS fine grains by using conical burr grinder, model DFJ-BL. Then, the rPS grains were sieved through a metal wire mesh with a mesh size of 5 mm. The size of rPS grains was kept under 5 mm to avoid big crumbs that might be taking more time to melt during the extrusion process and affect the quality of filament extruded. Figure 1 displays the process of rPS fine grains from the EPS wastes. The method of preparing rPS was referred to method reported by Ariel Leong et al. [1] and Ng. et al. [11]. The LDPE resin was purchased from Titan Plastic. LDPE resin and rPS fine grains were dried in oven for 6 hours at 60℃ to avoid any moisture causing air traps during the filament extrusion.

![Image](image_url)

Figure 1. Process of preparing rPS fine grains from Styrofoam wastes.

2.2. Production of Filament
The rPS/LDPE blends were formulated accordingly to the blend ratios of 90/10, 80/20, 70/30, 60/40, 50/50. For filament extrusion, the rPS grains and LDPE resin were pre-mixed manually before feeding into the machine. The rPS/LDPE mixture was fed into the single-screw Desktop Filament Extruder Line II (Wellzoom, China) with all heating zones’ temperatures set at 190℃. The extrusion speed was fixed at 40 rpm. The extruded filament went through a water bath that was controlled at room temperature to solidify the extruded filament. This equipment was not equipped with auto-calibration winding system. Hence, the diameter of the filament extruded was kept in the range of 1.5 – 1.75 mm by manually adjusting the speed of filament tractor. The winder was on when the filament diameter achieved desired range [1, 11]. Throughout the filament extruding process, the tractor speed and winder speed were monitored from time to time to keep filament diameter within desired range. Figure 2 illustrates the filament extrusion process.
2.3. 3D Printing of Specimens

For visual observation, the samples were printed with dimension of 20 mm × 20 mm × 2 mm. The dumbbell-shaped tensile specimens with dimension following ASTM D638 Type IV were printed for tensile test. The 3D models of sample and specimen were drawn using SolidWorks 2020 and saved in STL files. The STL files were sliced and converted into g-code files using Ultimaker Cura 4.8.0. Then, all specimens were printed using Creality Ender-3 V2 3D Printer with nozzle size 0.4 mm. Besides, the infill percentage and bed temperature were set at 100% and 110℃, respectively. All-purpose glue stick was also applied on the bed to promote better adhesion of the printed specimens on the bed to avoid warpage. Figure 3 displays the process flow of 3D printing the specimens.

The specimens were 3D printed using rPS/LDPE blend filament of all blend ratios. For visual observation, the samples were printed with different printing temperature and extrusion rate according to Table 1. For the tensile specimen, printing temperature and extrusion rate percentage were fixed at 240℃ and 120%, respectively.

| Type                      | Printing Temperature (℃) | Extrusion rate (%) |
|---------------------------|--------------------------|--------------------|
| Visual observation samples| 220, 230, 240            | 100, 110, 120      |
| Tensile testing specimens | 240                      | 120                |

Figure 2. Filament extrusion process.

Figure 3. Process flow of 3D printing the specimens.
2.4. Visual Observation
The visual observation was conducted using Swift M10 compound light microscope. The 4× object lens magnified images of the uppermost surfaces of the samples were taken using the software Motic Images Plus 3.0. The micrographs were recorded the image captured at crisscross printed region (spot A) and parallel printed wall (spot B) as labelled in Figure 4. The visual observation analysis was to study printing quality of specimen with variation of printing temperatures and extrusion rates.

![Figure 4](image)

Figure 4. Specimen for visual observation, A: crisscross printed region; B: parallel printed wall.

2.5. Tensile Testing
Tensile test was conducted to determine the tensile strength and tensile modulus of the printed tensile specimens. Instron’s Universal Testing Machine model 5969 equipped with the software Bluehill was used for the tensile test. A 15 kN loadcell and crosshead speed of 5 mm/min were used. More than 3 tensile specimens for each rPS/LDPE blend ratio were tested to get the average data.

2.6. Thermal Analysis
Thermogravimetric Analysis (TGA) was conducted to examine the thermal properties of different blend ratios of rPS/LDPE filaments. The filaments were sliced into small fragments with weight ranging from 8 to 10 mg. The fragments were placed into an alumina crucible. Perkin Elmer TGA 8000 was used in this experiment it was operated in the temperature range of 30 – 700°C using nitrogen gas with the flow rate of 20 ml/min and a heating rate of 10°C/min.

3. Results and Discussion
3.1. Visual Observation
Figure 5 displays the black spots that were observed in the printed layers of the specimens printed with rPS/LDPE blend filaments. The black spots that were embedded in printed layers were caused by the contaminants from rPS that obtained from EPS waste. Similar observation was also found by Ng et al. [11]. However, the presence of contaminants was not influencing the printing process.

The observations of the printing temperatures variation on the printing quality of printed specimens with rPS/LDPE blend filaments were shown in Figure 6. From the micrographs, the air gaps between printed layers of the specimens decreased when the printing temperature increased to 240°C. There were no air gaps found in the printed layers of the specimens printed at 240°C using rPS/LDPE filament with LDPE content more than 30 wt%. Printing temperature is one of the crucial aspects to control the melt viscosity of the molten rPS/LDPE filament. As reported by of Koay et al. [12], the reduction of viscosity of thermoplastic occurred with the increasing of process temperature due to the increment of polymer chain mobility. The entanglement of polymer chain and the intermolecular interaction were reduced. Thus, the flowability of material was increased.
Figure 5. Black spots occurred on the printed layers of rPS/LDPE blend specimen.

Regarding to Xu et al [13], the PS was naturally found to be having higher viscosity than LDPE, and the PS/LDPE blends’ viscosity would decrease with the increases of LDPE content. At printing temperature of 240°C, the air gaps were absence in the printed specimen using rPS/LDPE filament containing LDPE content at 30 wt% and beyond. When the viscosity of rPS/LDPE blends reduced due to the increase of printing temperature and LDPE content, the surface tension between printed layers were become smaller which resulting a good wettability occurred between the printed layers. Hence, the formation of coalescences on the printed layers would increase. The presence of more coalescences within the printed layers indicated the good adhesion behaviour between the printed layers. This finding shows the increase of printing temperature and LDPE content can enhance the adhesion between printed layers and achieve a good printing quality in the specimen. This statement also agreed by Singamneni et al. [14]. They agreed that using appropriate printing temperature for the material yielded suitable melt viscosity of the material that would encourage good adhesion and solidification between interlayer and inter stand of the printed samples.

Figure 6. Comparison of different printing temperature across the blend ratios of rPS/LDPE.
However, the increase of printing temperature was unable to guarantee a good printing quality on the printed specimen using rPS/LDPE filaments with different blend ratios. The air gaps were still visible for printed specimen using rPS/LDPE filaments with LDPE content lesser than 30 wt%. Besides, the diameter inconsistency of the self-extruded filament could also be one of the factors that might influence the adhesion of printed layers. The diameter of the commercial filament usually manufactured to be 1.75 mm with tolerance of 0.05 to ensure that the filament can feed into the hot end of the 3D printer. A small diameter tolerance of filament ensures the width consistency of printed layers that promotes good adhesion between printed layers. The rPS/LDPE blend filaments produced in this research have a higher tolerance in filament diameter as the equipment that used to extrude the filament was not equipped with auto-calibration winder system. To overcome this inconsistence issue, the extrusion rate of the 3D printing was manipulated.

As the increase of printing temperature could not entirely promote the adhesion between the printed layers for all rPS/LDPE blends of different blend ratios, the variation of extrusion rates was studied in this research. From Figure 7, the air gaps were significantly reduced when the extrusion rate percentage increased. The increase of extrusion rate percentage was attributed to more amount of molten rPS/LDPE filament being extruded during the print. As more amount of molten filament was being extruded, the width of the printed layers increased. When the width of the printed layers increased, the printed layers could form more coalescences and resulting a good adhesion between interlayer and inter stand of the printed specimen. According to Sukindar et al. [15], the increase of amount of the extruded molten filament led to a better printed layers’ adhesion in the printed specimen. At printing temperature of 240°C and extrusion rate of 120%, the rPS/LDPE blend filaments with different blend ratios exhibited absence of air gaps and good adhesion of printed layers in printed specimens. For this reason, the tensile specimens were printed with rPS/LDPE blend filaments using printing temperature of 240°C and extrusion rate of 120%.

| Extrusion Rate Percentage | 100% | 110% | 120% | 100% | 110% | 120% |
|---------------------------|------|------|------|------|------|------|
| A                         |      |      |      |      |      |      |
| B                         |      |      |      |      |      |      |

**Figure 7.** Comparison of different extrusion rate percentage across the blend ratios of rPS/LDPE.
3.2. Tensile Testing
For tensile test, the tensile properties of printed specimen with neat rPS filament were obtained from the study reported by Ng. et al. [11]. According to Figure 8, the tensile strength of neat rPS specimen was higher than the rPS/LDPE specimens with different blend ratios. The tensile strength of rPS/LDPE specimens were decreased the LDPE content in the blend increased. The rPS and LDPE blend is an immiscible polymer blend as the rPS and LDPE phases are incompatible. For the blend, the rPS and LDPE phases were unable to form a good interphase adhesion. Thus, this phenomenon might cause the tensile strength of the blend decreased. Identical finding was found on the report of PS/LDPE blends which was written by Barentsen & Heikens [16]. Besides, the incompatibility of rPS and LDPE also influenced the adhesion between printed layers. Although the printed layers were found to be having good interlayers’ adhesion and no air gaps in specimens regarding the finding from section 3.1, the strength of the adhesion between the printed layers might be weak due to the incompatibility between rPS and LDPE. Hence, the tensile strength of the specimens was decreased with increase of LDPE content in the blends. Seier et al. [17] also agreed that the compatibility of the polymers in the blend was critical in influencing the mechanical strength of the blend. They found that the incompatibility between two polymer phases significantly reduced the strength of the blend.

Figure 8. Tensile strength of 3D printed tensile specimen with rPS/LDPE blends.

Based on Figure 9, the tensile modulus of the neat rPS specimen was the lowest among rPS/LDPE specimens. Theoretically, the tensile modulus is reflected by the stiffness of the material which it shows the ratio of tensile stress over strain. In the experiment, the rPS/LDPE blend specimens exhibited low tensile strength but high tensile modulus. This was because the rPS/LDPE specimens were relatively brittle, and they fractured at a very low tensile strain. The brittleness of the rPS/LDPE specimens were caused by several causes which included (i) presence of weak adhesion strength between the printed layers and (ii) incompatibility between rPS and LDPE phases. From Figure 9, the tensile modulus of rPS/LDPE blend specimens were reduced when the LDPE content increased in the blend. As the LDPE was naturally more ductile than PS [16], the addition of more LDPE increased the overall ductility of the rPS/LDPE blend and caused the decreasing of tensile modulus.

The standard deviation error bars of the charts were noticeably large due to its high variation of data obtained. The large error bars might possibly cause by the impurities in the waste PS foam. Although cleaning process of the waste PS foam was conducted prior to the production of rPS/LDPE filaments, some of the impurities were still unavoidable as the EPS wastes were found porous and hard
to clean thoroughly. The impurities were acted as the stress concentrators that caused the initial crack to happen on the spot. The tensile properties of some specimens could be affected significantly when high number of impurities found in the specimens. Anderson [18] also found that the tensile properties of the printed specimen were found to be varied more as the specimens were printed using recycled material.

![Figure 9. Tensile modulus of 3D printed tensile specimen with rPS/LDPE blends.](image)

### 3.3. Thermal Analysis

The TGA curves of the neat rPS, neat LDPE and rPS/LDPE blends with different blend ratios are presented in Figure 10. The TGA curves of the neat rPS and neat LDPE had presented a single thermal degradation step while the TGA curves of rPS/LDPE blends had revealed two thermal degradation steps as highlighted in the red dotted circle. The two thermal degradation steps were occurred in the rPS/LDPE blends as the rPS phase in the blends were thermally degraded first and then followed by the thermal degradation of LDPE phase in the blends. The second thermal degradation steps of the rPS/LDPE blends became more noticeable when the content of LDPE increased. The onset thermal degradation temperatures ($T_{onset}$) obtained from the TGA curves were tabulated in Table 2. The $T_{onset}$ in TGA represents the temperature that the material starts to thermally degrade. Based on Table 2, the thermal resistance of LDPE was proven to be higher than rPS as the $T_{onset}$ of the neat LDPE was highest among the samples. The LDPE is semi crystalline polymer. The crystalline phase in LDPE is a highly compact polymer chains structure and it usually absorbs high amount of thermal energy before going through thermal degradation. In opposite, PS is naturally amorphous, the random arrangement of polymer chains in amorphous structure usually absorbs lesser thermal energy before degrading thermally. Thus, this was a common phenomenon when the LDPE had shown better thermal stability than rPS. Furthermore, the $T_{onset}$ of rPS/LDPE blends were shifted to higher temperature as the LDPE content in the blend increased. This indicated that the addition of LDPE contents in the rPS/LDPE blends enhanced its thermal resistance as compared to neat rPS. Ryu et al. [19] also found that the thermal stability of LDPE/PS blend was remarkably improved as compared to neat PS. For FDM printing process, filament with high thermal stability always a benefit as the dimension stability of a 3D printed object can be easily controlled.
Figure 10. TGA curves of neat rPS, neat LDPE and rPS/LDPE blends.

Table 2. Onset temperature obtained from TGA curves.

| Sample           | Onset Temperature (°C) |
|------------------|------------------------|
| neat rPS         | 407.5                  |
| rPS/LDPE 90/10   | 414.1                  |
| rPS/LDPE 80/20   | 421.5                  |
| rPS/LDPE 70/30   | 422.5                  |
| rPS/LDPE 60/40   | 424.8                  |
| rPS/LDPE 50/50   | 426.6                  |
| neat LDPE        | 470.2                  |

4. Conclusion
The result from the visual observation displayed that the rPS/LDPE sample revealed better adhesion between printed layers due to lower viscosity that was caused by higher printing temperature and higher LDPE content in rPS/LDPE blend. However, the change of the viscosity did not make rPS/LDPE specimens of all blend ratios printed with no air gaps. The addition of extrusion rate from 100% to 120% increased the extruded amount of the material and led to more coalescences between printer layers which promoted a better interlayer and inter stand layers in printed specimens. This research found that the rPS/LDPE filament with different blend ratios can be printed into 3D objects if the printing temperature set at 240°C and extrusion rate fixed at 120%. The tensile strengths of the rPS/LDPE specimens were lower than neat rPS specimens. The tensile strengths of rPS/LDPE specimens decreased when the LDPE content in the blend increased. The decrease of tensile strength of specimen mainly due to the incompatibility between rPS and LDPE. The tensile modulus of the rPS/LDPE specimens also decreased as the LDPE content in blend increased. The decrease of the tensile modulus in rPS/LDPE blend indicated that the ductility of the rPS/LDPE blend filament was increased due to the addition of more LDPE content. The addition of more LDPE content in the blend also increased the thermal ability of rPS/LDPE blends as compared to neat rPS. This research demonstrated the potential of producing polymer blend filament from the EPS waste. The rPS/LDPE blend filament can be printed into a 3D object. However, the rPS/LDPE blend filament could be further improved if it adds with compatibilizer.
References

[1] Ariel Leong J J, Koay S C, Chan M Y, Choo H L, Tshai K Y, and Ong T K 2021 J. Nat. Fibres In Press doi: 10.1080/15440478.2021.1941488
[2] Conner B P, Manogharan G P, Martof A N, Rodomsky L M, Rodomsky C M, Jordan D C and Limperos J W 2014 Addit. Manuf. 1-4 64
[3] Bankole I O, Sikiru O I, Temitope D A, David B O, and Mohsen Z 2021 Mater. Chem. Phys. 258 123943
[4] Juarez C Z J, Koay S C, Chan M Y, Choo H L, Pang M M and Ong T K 2021 MATEC Web Conf. 335 03007
[5] Ngo T D, Kashani A, Imbalzano G, Nguyen K T Q and Hui D 2018 Compos. Part B Eng. 143 172
[6] Ong T K, Choo H L, Choo W J, Koay S C and Pang M M 2020 Adv. Manuf. Eng. 725.
[7] Chun K S, Fahamy N M Y, Yeng C Y, Choo H L, Pang M M and Tshai K Y 2018 J. Eng. Sci. Technol. 13 3445.
[8] Koay S C, Subramanian V, Chan M Y, Pang M M, Tshai K Y and Cheah K H 2018 MATEC Web Conf. 152 02019
[9] Ling S L, Koay S C, Chan M Y, Tshai K Y, Chantara T R and Pang M M 2020 Polym. Eng. Sci. 60 202
[10] Chun K S, Subramaniam V, Yeng C M, Meng P M, Ratnam C T, Yeow T K and How C K 2019 J. Thermoplast. Compos. Mater. 32 1455
[11] Ng T Y, Koay S C, Chan M Y, Choo H L and Ong T K 2020 AIP Conf. Proc. 2233 020022
[12] Koay S C, Husseinsyah S and Yeng C M 2017 J. Thermoplast. Compos. Mater. 30 1217
[13] Xu S A, Zhu L, Xie J W and Jiang M 1999 Polym. Int. 48 1113
[14] Singamneni S, Smith D, LeGuen M J and Truong D 2018 Polym. 10 922
[15] Sukindar N A, Ariffin M K A, Hang Tuah Baharudin B T, Jaafar C N A and Ismail M I S 2016 J. Teknol. 78
[16] Barentsen W M and Heikens D 1973 Polym. 14 579
[17] Seier M, Stanic S, Koch T and Archodoulaki V M 2020 Polym. 12 1
[18] Anderson I 2017 3D Print. Addit. Manuf. 4 110
[19] Ryu J G, Kim H, Kim M H and Lee J W 2004 Korea Aust. Rheol. J. 16 147