Solid Freeform Fabrication 2017: Proceedings of the 28th Annual International
Solid Freeform Fabrication Symposium – An Additive Manufacturing Conference
Reviewed Paper

EcoPrinting: Investigating the use of 100% recycled Acrylonitrile Butadiene Styrene (ABS) for Additive Manufacturing

Mazher Iqbal Mohammed, Anirudra Das, Eli Gomez-Kervin, Daniel Wilson, Ian Gibson

Deakin University, School of Engineering, 75 Pigdons Road, Waurn Ponds, Geelong, VIC 3216, Australia

Keywords: 3D Printing, Recycling, FDM, EcoPrinting, ABS

Corresponding Author: mazher.mohammed@deakin.edu.au

Abstract

Many commonly found polymers have the potential to be recycled, such as Acrylonitrile Butadiene Styrene (ABS), a prevalent 3D printing material. In this study we examine the potential of using 100% recycled ABS to form filaments for use in Fused Deposition Modelling (FDM) 3D printing. We then characterise the resulting changes in the printing quality and mechanical properties, over a single recycling cycle. We found that ABS can undergo recycling and reforming into consistent printer filaments without the addition of virgin material. However, notable changes in polymer characteristics were observed, reflected by degradation in mechanical properties during tensile tests and a decrease in the polymer melt flow, which required reduced raster speed to achieve repeatable prints. Despite these limitations, we demonstrate that recycling and reprinting is possible with acceptable loss of material integrity, and could provide unique opportunities for sustainable use of waste ABS using 3D printing technology.

1 Introduction

Plastics are a highly pervasive and integral part of our daily life, found in everything from packaging, furniture, automobiles, micro devices and even biomedical components [1-5]. The prevalence of plastics can be attributed to their highly desirable material properties, such as their lightweight, durability and amenability to be manufacturing using a diverse range of standardised processes, such as casting, CNC machining [6], laser ablation/engraving [2], injection moulding [7] and 3D printing [5, 8]. The usefulness of plastic and the optimisation of its use in manufacturing has resulted in an escalation in its global production, rising by over 500% in the last three decades, and is set to increase to 850 million tons annually by the year 2050 [9]. This intensive production and use of plastics is resulting to several unprecedented issues relating to the disposal and recycling of this material given the sheer volume that is to be processed. Additionally, most plastics are not biodegradable and produced from oil. Therefore plastic recycling is becoming increasingly important, not only due to finite fossil based resources on our earth which are simultaneously used for energy generation, but from the resulting negative impact our current actions are have on the environment.

Arguably, sustainable management of plastics has now become one of the biggest challenge we now face in the modern day.

In an ideal scenario, the lowest impact on the environment would be the direct re-use of a plastic products and devices. However, this may not always be practical should a product become compromised or damaged beyond the effective usefulness. Additionally, in the case of typical plastic FDM printing, a significant volume of material is lost due to the use of rafts, supports and failed prints, leaving such volumes of material typically redundant. In such scenarios recycling becomes the most attractive option to make best use of this finite resource.

With respect to additive manufacturing, plastics arguable comprise the most widely used feedstock for 3D printers and have become ubiquitous in our daily lives through the popularity and accessibility of polymer FDM printers. FDM systems operate by the use of thermoformable plastic filaments which are progressively fed into a heated nozzle, and the liquefied material is extruded outwards to form the respective layers of a printed object [10]. In essence the filaments used in FDM printers are identical to the filaments produced during the extrusion phase of the classical industrial recycling process. Therefore we believed that using these same processes with waste recyclable plastics, otherwise destined for landfill, we can generate filaments for use in FDM based 3D printers. Such concepts are rapidly becoming of research interest to many groups worldwide [9, 11-13].

In this study we investigate the reformation of 100% recycled 3D printed ABS material, into 3D printer filaments and assess the printing quality and integrity of this as a primary FDM feedstock. We investigate the parameters relating to the production of FDM filament using both virgin pelletised and recycled granules and characterise the resulting filament diameter and melt flow rate for varying extrusion temperatures. Using the most promising filament extrusion parameters and resulting filament, we examine the print quality in terms of layer adhesion and surface roughness of the printed material. We also perform tensile testing of printed samples to assess material degradation resulting from the recycling process. Our initial findings confirm that repeatable FDM 3D printer filaments can be produced using 100% recycled ABS and used to print parts with a similar build quality to commercial printer filaments, yielding near equal surface roughness profiles. We found there was a marked decrease of 13 to 49% in the ultimate strength of several test samples, depending on the print orientation. Overall, we have found that the superficial print quality and ultimate strength to be acceptable for most printing applications, leading us to believe that recycling of ABS plastics could provide a viable means of upcycling waste 3D printed ABS material. This technique could help both research and home users obtain greater longevity from 3D printer feed stocks and potentially reduce the escalating burden on landfill.

2 Methodology

2.1 Plastic Granulation

ABS plastic was obtain from either the use of virgin pellet material (Filastruder, GA, USA) or from failed/redundant 3D prints and raft/support material from teaching classes and research use within the School of Engineering at Deakin University. No restrictions were imposed to ensure that the recycled ABS can from a single filament supplier source or ABS grade, with the exception of the ABS pellets, which were all sourced from the single batch. This generic use of ABS for recycling purposes ensured that there was no bias in the results, whereby plastics containing specific plasticisers or additives which could enhance in the recycling process were utilised. Therefore our results are likely to be reflective of general conditions experienced when recycling batches of ABS for wider reproducibility of our findings.

Waste ABS was granulated using a combination of crushing in a bench vice, direct cutting using wire cutters and the use of modified paper shredder. The bench vice was used crush larger printed models into smaller pieces (approximately >8cm^3) by clamping and applying torque to compress the respective model. The wire cutters were used to perform finer size reductions, before the resulting pieced were fed into the modified paper shredder to produce granules of ABS. To ensure a relatively uniform size of granule, pieces were separated using a sieve with a mesh size of approximately 5mm.

2.2 Filament generation

To produce the 3D printer filaments, both the pelletised material and the granulated ABS was processed through a commercial filament extruder (Filabot Ex2, Barre VT, USA). The extruder operates by the loading of plastic material into a hopper, which then using a drive screw, delivers material through a temperature controlled heated region. At sufficient temperature, the plastic softens and liquefies before being forced out of the device through a nozzle of a fixed diameter due to growing pressure from the continuous addition of plastic material. The diameter of the resulting filament can be controlled based on the temperature of the heating zone, the rotational speed of the drive screw and the size of the granules placed into the hopper and the nozzle diameter. The temperature effectively controls the viscosity of the loaded ABS once it enters its molten state, while the drive screw rotational speed and granule size influence the feed rate, which in turn dictates the pressure applied to the molten material for extrusion. Combined, these factors allow for control of the ABS melt flow rate out of a fixed size extruder nozzle, which then dictates the resulting diameter of the extruded filament. By careful control of all of these parameters, we are able control the filament diameter to obtain filaments of a suitable size for use in the 3D printers. The printer employed in this study was designed to accept filaments with either a diameter of 1.75mm or 2.85mm and could readily accept filament diameters between this ranges. For this initial study, we opted to produce 1.75mm filaments. Owing to the lack of any robust spooling equipment, to generate the filaments we utilised a simple gravity extrusion based methodology. In this process the extruded material was allowed to flow from the nozzle, at a height of 860mm, and gradually make its way to the floor under its own weight. We found that this methodology worked effectively for the use of ABS to realise a simple solution for continuous production.

Filaments were generated using virgin pelletised ABS and 100% recycling ABS. For each variant of ABS, care was taken to ensure a relatively uniform granule size. When producing the filament, the extrusion speed was fixed at 50% (approximately 17.5rpm), the nozzle diameter at 1.75mm, and the temperature adjusted accordingly to obtain filaments of approximately 1.75mm. Once filament production had reached a steady state, for each temperature and ABS variant we recorded the length of material that was extruded over a one minute time period to estimate the melt flow rate of the polymer.

2.3 3D printing and characterisation

All 3D printing in this study was performed using a commercially available FDM printer (Flashforge Dreamer, USA), which utilises Flashprint proprietary software interface and allows for control of all printing parameters (extrusion temperature, feed rate, etc) to optimise the final models. The printer also has a heated bed to allow for improved adhesion of ABS during the building process. For this study, the virgin filament material was printed using standard printing parameters, comprising

235^{o}C nozzle temperature and 105^{o}C bed temperature respectively. For the recycled material, the bed temperature was held constant unless issues with adhesion were observed, and other influential printing parameters were adjusted stepwise until optimal model production was achieved (no gaps, even layer adhesion, etc).

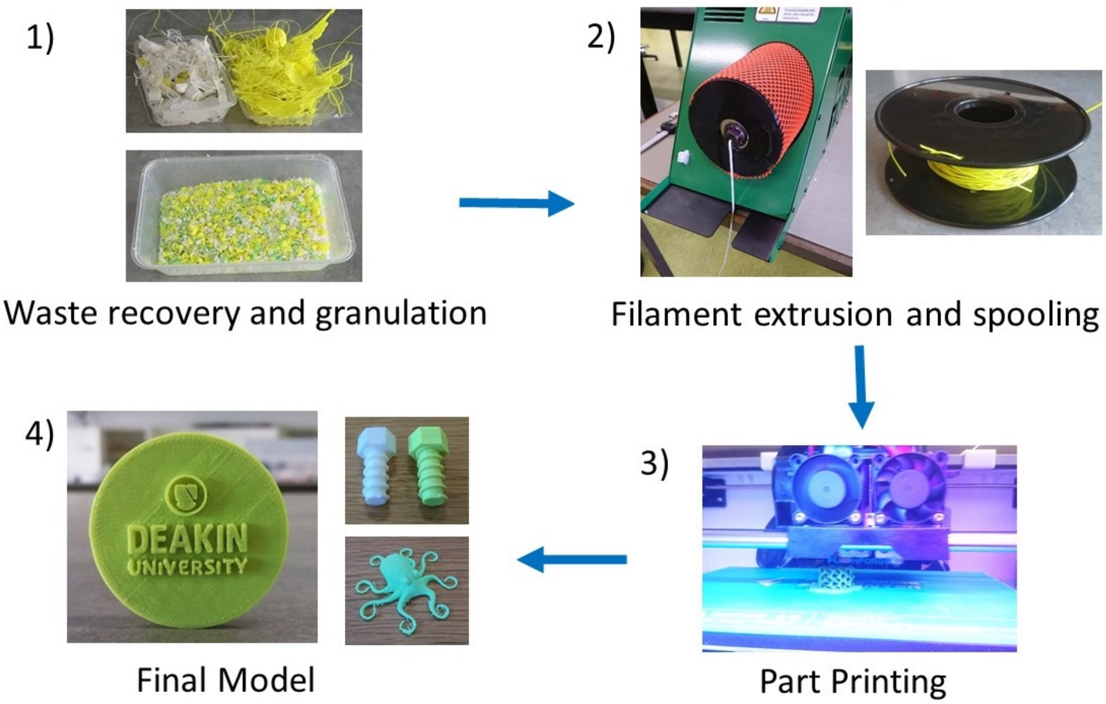

Figure 01: An overview of the ABS recycling process, comprising 1) ABS recovery and granulation 2) filament extrusion and spooling, 3) FDM 3D printing and 4) the resulting final models.

To examine the material properties of the recycled ABS, as compared to virgin ABS, various test specimens for tensile tests were printed accordance to ISO 527-2 European standards for plastics [14]. Given limited availability of recyclable ABS plastics for this study and in keeping in the spirit of the sustainable credentials of this study, test specimens were made to the type 1BA standard, which minimised overall material consumption. Tensile tests were performed using an Instron 5960 Dual Column 50kN tensile test system (MA, USA), where five repeat measurements were conducted. Only results which conformed to the standardised test procedure were recorded and all other samples were discarded until five measurements were achieved. Beyond tensile testing, various test models were fabricated to qualitatively examine the printability of the recycled filaments in realistic scenarios, for ‘passive’ (models) and ‘active’ parts (nut and bolt) to demonstrate the viability of using 100% recycled ABS material.

3 Results

3.1 Granulation

With respect to commercial filament generation, the industry standard comprises the processing of highly uniform pelletised material in a melt extrusion process. We adopt a similar methodology for the generation of recycled plastic filaments, however make uses of coarse granulated material, attained by repeated grinding in the modified paper shredder. Twenty virgin ABS pellets were randomly collected and their diameters measured. The average pellet size was determined to be approximately 3.4±0.4mm, and ranged across diameters of approximately 3-4.5mm. This value formed the baseline average we would aim to achieve with the granulated recycled ABS material, thereby ensuring that this variables influence on the polymer feed rate was kept uniform between the recycled and commercial ABS.

Typically, the waste plastic required 4-5 repeated shredding cycles and processing through the 5mm mesh sieve to achieve suitable granules. Figure 1 shows a sample of the granulated material obtained from the shredding and sieving process. Twenty randomly selected granules were measured for their average diameter. The average granule size was determined to be approximately 3.8±1mm, and ranged across diameters/maximum lengths from between 2.3-6.3mm, in the examined samples. It was found that despite being generally effective, the current methodology did at times allow for larger granules to pass through the sieve. Standard methods for palletisation of the recycled material would have undoubtedly increased the granule diameter consistency. In this study we opted against this approach as to do so would result in potential thermal degradation of the recycled polymer material, which in turn would influence the subsequent mechanical tests. Ultimately, the obtained average would allow for a relatively uniform material feed rate into the extruder.

3.2 Filaments

To produce the filaments, either the pelletised or granulated plastics were loaded into the hopper of the extruder and fed into the heating barrel for production at a fixed extrusion speed of approximately 17.5rpm. From preliminary tests we established that extrusion was possible at approximately 180^{o}C, however showed signs of containing coarse polymer material. This temperature was used as a baseline lower limit for extrusion and working in steps of approximately 5^{o}C, the average diameter of the filament was measured up to a maximum of 220^{o}C. For each temperature setting the melt flow characteristics of the polymer were realised.

It was found that there was a measureable difference in the filament diameter until the extrusion process has reached a steady state (with the material touching the floor and coiling). We found that there was an approximate difference of approximately up to 0.4mm in the measured diameter, and believe to be due to the gravitational forces of the filament stretching under the weight of the previously extruded material. Once the steady state had been reached, the gravitational force remained relatively constant due to an approximate equal mass of material being between the nozzle and floor, resulting in a relatively uniform filament diameter for further extrusion. At this point a measured maximum deviation of 0.15mm was found from the average filament diameter.

Figure 2bi) illustrates graphs of the change in extruder filament diameter and the average extrusion speed, for changes in the temperature for both the pelletised and 100% recycled ABS material. With respect to filament diameter, it can be seen that both ABS variants show a decrease in the average filament diameter for an increase in the extrusion temperature. This would be expected as at the start temperature of 185^{o}C, which is well above the glass transition temperature of ABS (≈105^{o}C), the polymer is in a liquid phase state. Therefore increases in temperature will result in a decreasing the relative viscosity of the bulk polymer, resulting in greater melt flow through the nozzle (as observed in figure 2bii), and hence a decrease in the overall extrusion diameter. Interestingly, for 100% recycled ABS, a relatively uniform melt flow rate from the extruder was observed, with a modest rise of approximately 5-6mm^3/s from temperatures of 185 to 205°C, as can be seen in figure 2bii). This discrepancy could be explained by the irregularities in the average granule size of the recycled ABS, which results in unpredictable fluctuations in the feed rate of the ABS into the extrusion nozzle, and hence sporadic changes in the melt flow rate. This thought is substantiated by the relatively uniform increase in melt flow rate for the pelletised ABS, which has a much greater uniformity in average pellet size, and hence more predictable feed rate characteristics. However, there is exists a possibility that the relatively uniform melt flow rate could be due to some form of degradation of the ABS material. To test this assumption, should we perform prints with increasing nozzle temperature, we should expect the extrusion volume to remain relatively constant. We therefore conducted test prints varying the nozzle temperature to check this hypothesis, as detailed in section 3.3.

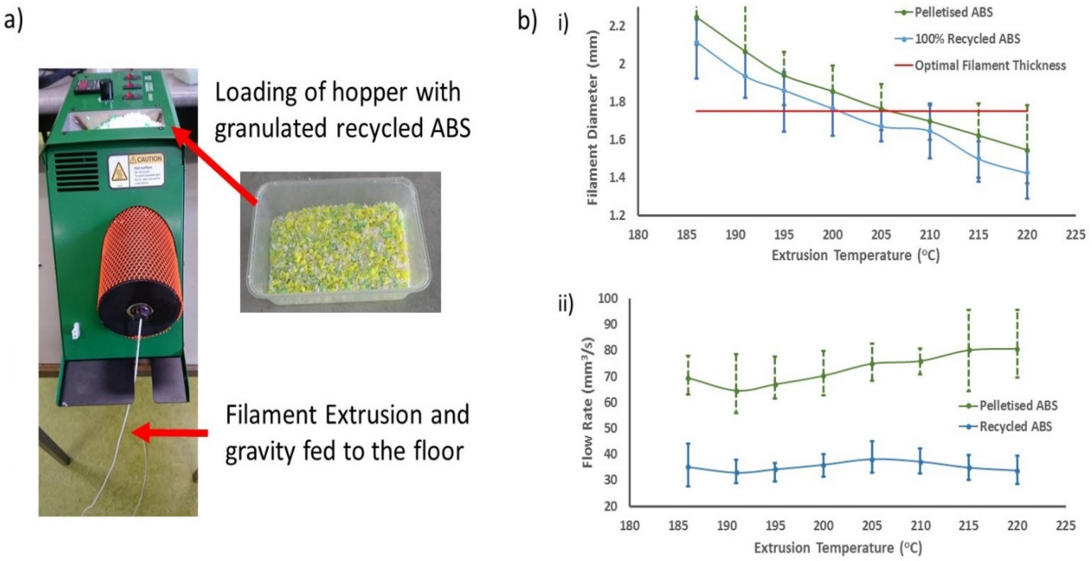

Figure 02: a) Illustration of the recycling melt extrusion process, whereby the recycled ABS granules are loaded into the extruder and the resulting filament is gravity fed to the floor. b) Graphs illustrating the results for i) change in filament diameter for increased extrusion temperature and ii) Change in melt flow rate for increased extrusion temperature.

Overall, examining the results in figure 2bi), the pelletised virgin and 100% recycled ABS show similar rates of diameter decrease for a respective rise in temperature, excluding the spurious result at 210°C. It was found that to obtain a filament diameters of 1.75mm, the virgin pelletised ABS was to be extruded at approximately 205°C and the recycled ABS at 200°C respectively. Speculating, this decrease in the extrusion temperature may be indicative of a decrease in the mechanical integrity of the ABS resulting from thermo-oxidative degradation, as has been described previously [15, 16]. Using the resulting extrusion temperatures filaments of 1.76±0.12mm we produced using the pelletised ABS and 1.76±0.14mm for the recycled ABS respectively for subsequent mechanical testing and qualitative printing analysis.

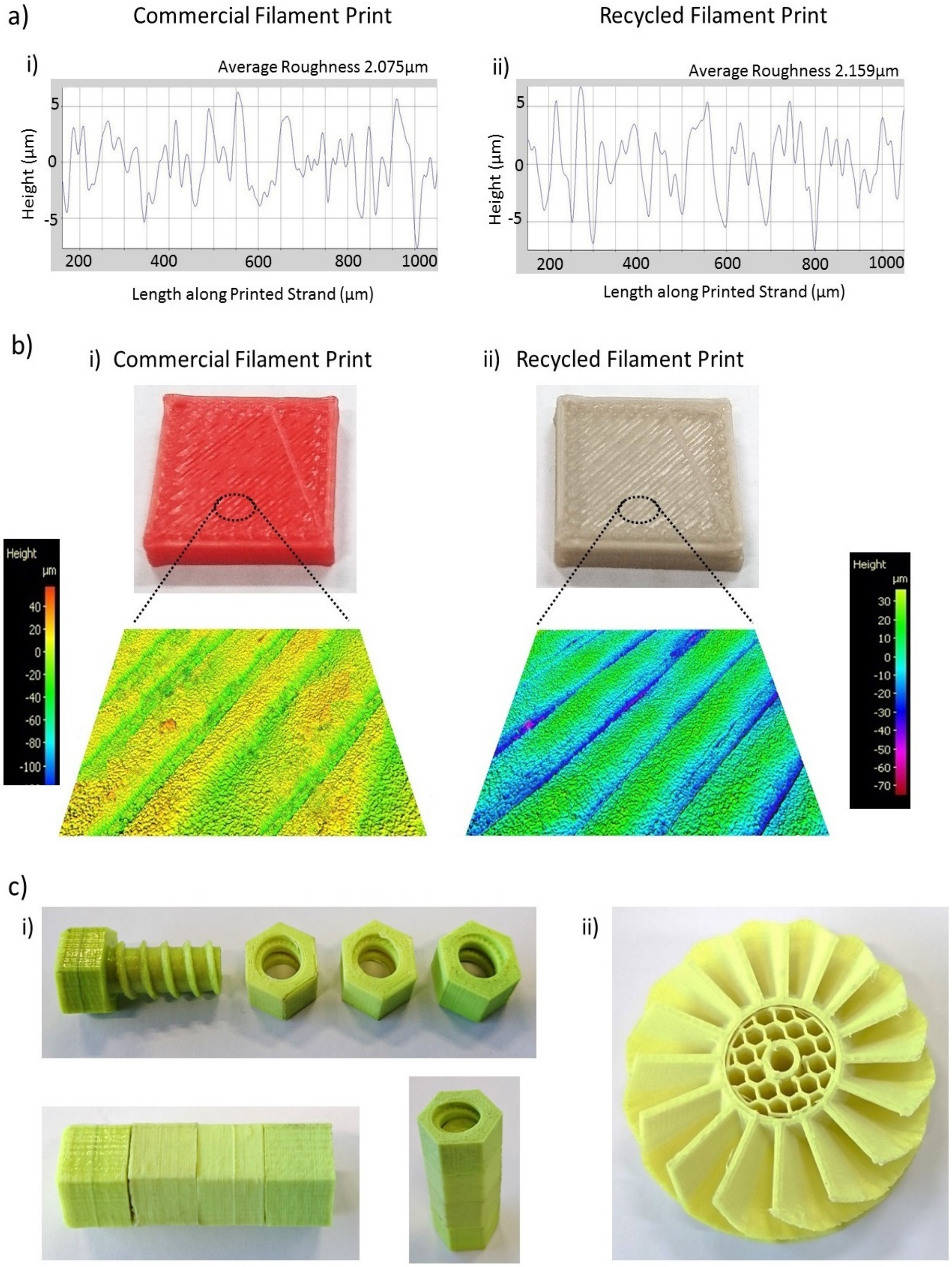

Figure 03: a) Graphs illustrating the surface roughness of printed section of a square test print using both commercial and recycled 3D printer filaments. b) Pictures of the square test print and surface topography profiles when using i) commercial and ii) recycling 3D printer filaments. c) Test print structures from the recycled ABS filaments comprising i) a bolt and 3 nuts and ii) a propeller concept.

3.3 3D printing

Upon formation of the filament from each respective ABS variant, the resulting material was loaded into the 3D printer for testing. Examining the filament produced using the pelletised ABS, it was found that the printing performance matched that of standard commercial ABS filaments, with optimal printing parameters comprising nozzle temperature 235^{o}C, print speed of 60mm/s and bed temperature of 105^{o}C. When the recycled ABS was printed at these same parameters, it was found that the resulting prints showed signs of under extrusion in the form of gaps in the extruded strands of the print (results not show). Typically, to remedy this issue using commercial filaments, one can raise the nozzle temperature, which results in an increase in the melt flow rate of the ABS and a greater volume of deposited material. As we recorded a relatively uniform melt flow rate for the recycled material, an increase in the nozzle temperature would not result in an increase in the deposited material. This was confirmed by some preliminary printing tests examining nozzle temperatures of 240, 245 and 250^{o}C. In this instance to compensate for the under extrusion, the printing speed was reduced, with optimal printing parameters comprising nozzle temperature 235^{o}C, print speed of 50mm/s and bed temperature of 105^{o}C. It is also speculated that we could equally increase the filament feed rate to compensate for the under extrusion, and we hope to examine this in future studies. Ultimately, we achieved reliable printing parameters to achieve consistent printed models and so began to scrutinise parameters relating to the printing quality and material integrity.

3.3.1 Print quality tests

To examine the printability of the recycled material in comparison to virgin material we printed a test structure for comparative purposes, comprising a square of 20x20x5mm, and performed various surface profile measurements. Figure 3 illustrates the final test structures and the resulting 3D topography scans, alongside data for the surface roughness over a region randomly selected region following the direction of the deposited strand. It was found that the average deviation in the surface was relatively uniform between the virgin and recycled ABS material, with an approximate deviation of 50µm between the peaks of trough of where the deposited ABS material join. Equally the average surface roughness of the deposited material was found to be similar, with an average roughness along the length of a deposited ABS stand of 2.08µm for commercial filament and 2.16µm for recycled filaments respectively. These test would indicate a similar print quality between the commercial and recycled filaments for the derived printing parameters. To qualitatively demonstrate the print quality of the recycled filament, we performed test prints of designed engineering items, comprising a simple nut and bolt system and a propeller concept. Both items were challenging prints for even commercial grade filaments and were printed successfully in recycled material on our first attempts, as illustrated in figure 3c).

3.3.2 Tensile tests

As we had now established that the printing quality of the recycled filaments could match the performance of commercial filament, we examined the changes in the mechanical properties of the printed material as a metric to examine material degradation. Tensile tests were performed by clamping a test specimen in the Instron test rig (figure 4ai), while a constant load was applied beyond initial elongation and on to failure. Samples were printed all three potential print orientations, as seen in figure 4aii), using a print infill of 100%. Sample results from the tensile tests can be seen in the graphs in figure 4b), which illustrates a comparison between the commercial and recycled printed specimens for samples manufactured in the X build orientation. A summary of the complete test data can be seen in table 1.

It was found that there was minimal difference in the ultimate strength and average stiffness for use of the virgin ABS filament in the X and Y build orientations, and an approximate 25% comparative decrease for samples build in the Z orientation. By comparison there was a clear decrease in the recorded ultimate strength and average stiffness for the recycled ABS, across all build orientations, with the X build orientation offering the greatest level of mechanical strength and rigidity. Comparing the ultimate strength between the virgin and recycled filaments, it can be seen there is a clear decrease of between 13 to 49%, with the X build orientation demonstrating the lowest percentage decrease (13%). Equally, there was a marked decrease in the material stiffness with measured decreases of between 17 to 28%, with again the X build orientation demonstrating the lowest percentage decrease (17%). Clearly these results are an indication of the mechanical degradation of the ABS plastic during recycling, but also hint that the build integrity of the test sample (e.g. layer adhesion) is also compromised for the given parameters utilised. Should this be the case, it would explain why the results for the ultimate strength would be greatest for the X build orientation and lowest for the Z build orientations respectively given the translational mode of the printer nozzle during the polymer extrusion process.

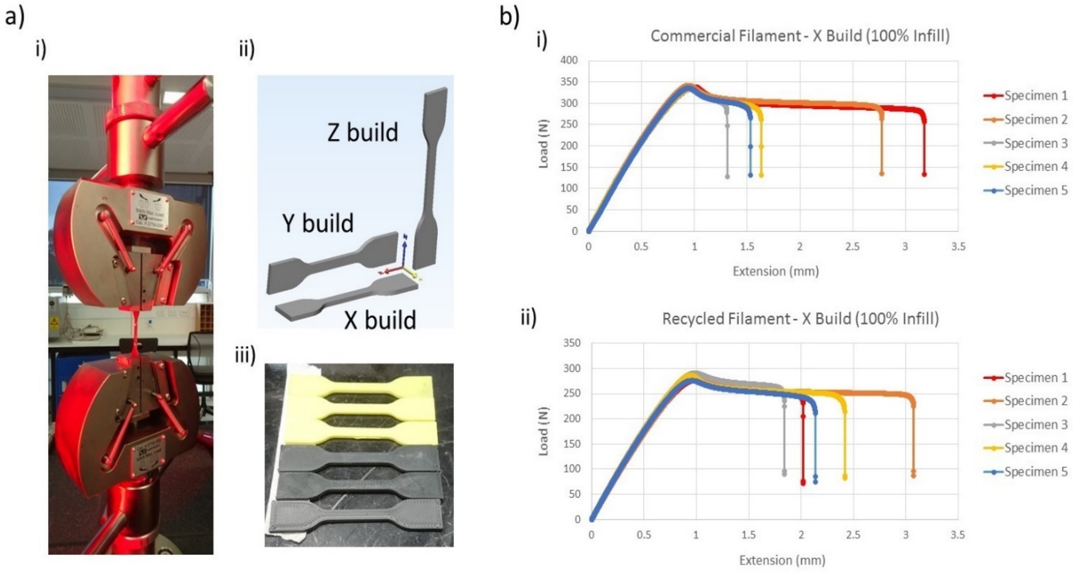

Figure 04: a) i) Photograph of the tensile test apparatus, ii) Diagram of the examined build orientations for the printed dog bone samples and iii) printed dog bone samples awaiting testing. b) Graphs depicting the results of tensile tests for dog bones built in the X build orientation and 100% infill for i) commercial and ii) recycling 3D printer filaments.

In terms of the strain failure of the test samples, there was marginal difference between the commercial and recycled samples, with the exception of the samples in the z orientation, which showed an approximate 50% decrease. We believe these results are again indicative of the compromised layer adhesion of the recycled material, which would be exacerbated for samples manufactured in the Z orientation. Indeed, it was visually observed that there was compromised layer adhesion on the Z build samples prior to tensile tests.

Results ultimately confirm that there is degradation in the mechanical properties of the recycled material, but also that there may be issues with the manufacturing process and the sample type utilised. In a recent study by Torrado et al 2016, varying ultimate tensile strength (UTS) was observed between samples built in differing ASTM standard tensile types, rastering direction and orientation [17]. There findings would perhaps indicate the there are issues with relying on FDM printed samples built vertically (Z build orientation) to determine the UTS, particularly between the varying size types as part of recognised standards. We believe their study highlights the necessity to validate our claims in this study with further tests using tensile samples of increased size and control of the rastering direction. We hope to perform further tests in future continuations of this study.

	Commercial Filament			Recycled Filament		
Sample	Ultimate Strength (MPa)	Average Stiffness (N/mm)	Strain at Failure (%)	Ultimate Strength (MPa)	Average Stiffness (N/mm)	Strain at Failure (%)
X Build	40.59±1.42	430.50	10.35±1.73	35.44±2.01	357.40	11.62±1.13
Y Build	42.34±0.88	419.78	6.62±0.28	29.06±1.21	302	6.36±0.18
Z Build	33.25±0.66	404.05	4.62±0.51	16.81±0.66	329.80	2.47±0.23

Table 01: A summary of the tensile tests for each of the examined printing orientations using both commercial and recycled filaments.

4 Conclusions

In this study we have demonstrated a methodology for the reforming of 100% waste ABS plastics into relatively uniform 3D printer filaments (1.76±0.14mm). When forming the filaments we found that increasing the extrusion temperature reduced the extruded filament diameter through the extruder die. Interestingly, the melt flow rate of the recycled material remained relatively uniform for increased extrusion temperature, whereas virgin pelletised ABS showed an increase in melt flow rate. This was suspected to be due to thermal degradation of the recycled ABS, and required the adjustment of the raster speed to avoid under extrusion when printing. It was found that the superficial print quality of the recycled filament was of a similar consistency as commercial filaments, as confirmed by surface roughness analysis. However there was a marked degradation in the mechanical properties of the recycled ABS, resulting in a 13 to 49% decrease in the ultimate strength of the printed material. Ultimately, we believe that our methodology to extrude 100% recycled ABS plastic to be a viable option to make better use of printed resources, and could prove useful in the management of other sources of waste ABS, in addition to alternative recyclable plastics.

5 References

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