

Waste to filaments: Turning ocean waste into filaments for extrusion-based additive manufacturing

F. Grønborg^{1,2} and D. B. Pedersen¹

¹Technical University of Denmark, Denmark

²Bjørn Thorsen A/S, Denmark

FRGMA@dtu.dk

Abstract

Waste plastics are generally considered to have limited value and are often regarded as an expenditure. It is feasible to process the majority of plastic wastes into filaments for Extrusion-based Additive Manufacturing, yet their suitability must be tested. The recycling process and the implementation of an Extrusion-based Additive Manufacturing strategy are crucial for testing the suitability for production of usable parts. This study focuses on utilizing Twin Screw Extrusion operating at moderately high screw speeds, employing compatibilization techniques, and utilizing maleic anhydride grafted thermoplastic elastomers to generate filaments from ocean waste. The printability and part design strategies are assessed to identify effective approaches for incorporating plastic waste into a comprehensive Additive Manufacturing process.

Extrusion-based Additive Manufacturing, Ocean Waste, Filaments, Warp-index

1. Introduction

Plastic pollution in the oceans poses an escalating challenge, with its complete implications yet to be fully understood. A considerable portion of this waste eventually washes ashore, where it is often collected by volunteers, beachgoers, and, in this case, school children in collaboration with the small Danish company called *Strandet Aps*. Nonetheless, the mixed composition and degraded quality of the collected plastic restrict its practicality, often necessitating downcycling for energy recovery rather than repurposing.

Utilizing plastic beach waste as primary constituents for Extrusion-Based Additive Manufacturing (EBAM) proves particularly challenging. The predominant polymers found in this oceanic waste are polyethylene (PE) and polypropylene (PP), which are not only widely used but also buoyant, thereby significantly contributing to marine debris. PE and PP are known

to exhibit high shrinkage, making them less desirable for use as filaments in EBAM processes.

The impact of key parameters on the fabrication process and the properties of the parts is well understood. The motivation behind utilizing ocean waste is twofold: (1) to advance the knowledge of printing with recycled and highly degraded polymer materials, and (2) to explore alternative applications for ocean plastic waste that do not involve energy recovery. Extensive documentation exists regarding the influence of different parameters on the fabrication process and the resulting part properties.

To address warpage issues, techniques such as employing higher nozzle temperatures, slower printing speeds, and thinner layers have been proposed [1], [2]. The challenge posed by the adhesive properties of polyolefinic polymers like PE and PP lies in the interdiffusion and entanglement of polymer chains across the layers [3]. Striking a balance between sufficient energy for

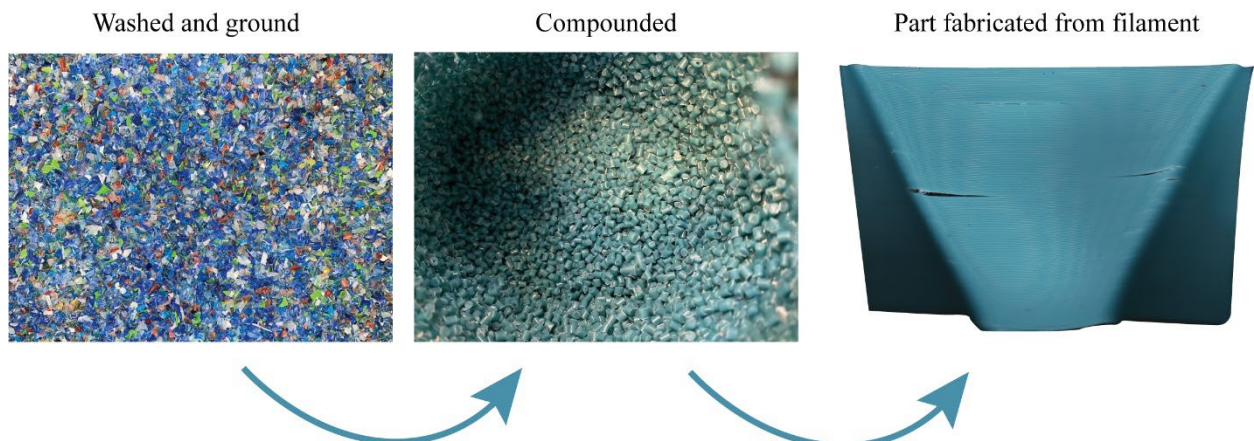


Figure 1. The stages from waste to print. The washed and ground flakes with mixed, but mainly blue colours are giving the pellets a turquoise colour

polymer entanglement without compromising the part's mechanical integrity during printing is a key concern. Disentanglement occurs within the nozzle due to high shear flow, leading to a recovery time that reduces interlayer bonding [3]. Increasing the temperature has been shown to enhance tear strength between layers [4], aligning with the concept of providing more energy for entanglement. Enhancing interlayer adhesion can also be achieved by increasing the contact area through the reduction of layer height and implementing "over-extrusion" [5].

On the material side, warping is primarily influenced by the coefficient of thermal expansion (CTE) [6]. The addition of fillers with lower CTE can help mitigate warping; however, this approach may increase the complexity and cost of recycling the material in the future. Reactive compatibilizers have demonstrated the potential to improve strength in PP/PET compounds [7], which could theoretically enhance interlayer bonding. Exploring the usability of reactive polymer compatibilizers for EBAM and identifying potential applications for such materials remain areas of interest.

2. Method and Materials

Plastic waste was collected from beaches on the west coast of Jutland, Denmark by Strandet Aps. The collected waste underwent cleaning and shredding before compounding. Compounding of the waste plastic, along with an additional compatibilizer, was carried out using a Thermofisher Eurolab 16XL. Without the presence of the compatibilizer, the material did not extrude to a usable quality. Two compound variations were prepared, consisting of 95/5 wt% and 90/10 wt% ocean waste and Acti-Tech 18MA08 (AT). During the processing of the compounds, a flat temperature profile of 200°C and 300 RPM at a rate of 1 kg/h was maintained.

Dynamic Scanning Calorimetry (DSC) measurements were conducted using a TA Instruments Discovery DSC. The measurements involved a temperature ramp of 10.00°C/min up to 250.00°C, with a 5.00-minute isothermal rest before and after each ramp cycle.

Tensile tests were performed on both filaments and manufactured tensile specimens using the Mecmesin Multitest 2.5-l machine, applying a speed of 50 mm/min. Tensile bars of S2 type, fabricated using the produced filaments, served as the test specimens.



Figure 2. The warp testing specimen.

To assess the warping of the material, a test part was designed based on Parthy's work in 2015 [8]. Evaluation of warping was carried out by fabricating a warp test specimen seen in Figure 2 using an enclosed direct drive EBAM machine, while varying the printing parameters. This design allows for relatively free warping of the layers. Parthy suggests evaluating the curvature of the layers by utilizing a flatbed scanner to create a warp index, measuring the height of the warped layer, and dividing it by its width, as illustrated in Figure 3.

$$\text{Warp index} = \frac{\text{Height of warp}}{\text{Width of layer}}$$

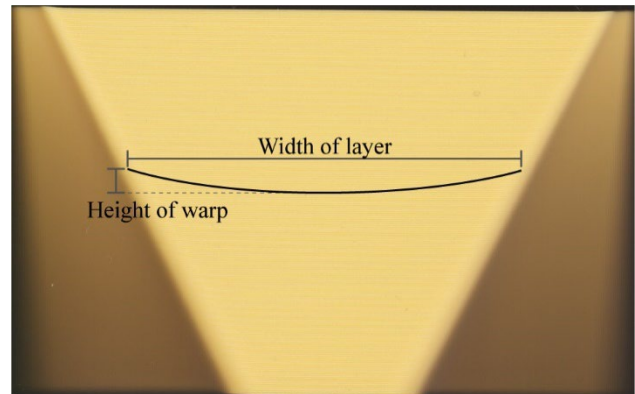


Figure 3. PLA benchmark print with an exaggerated warp line to illustrate the warp index.

3. Results and Discussion

Upon receiving the materials, they were initially extruded without any additives, resulting in an inhomogeneous extrudate with insufficient melt strength to form a filament. However, after the addition of 5 or 10 wt% compatibilizer, the melt strength was restored, allowing for filament formation. This preliminary experiment indicates that the plastic waste received had undergone significant degradation due to its exposure to the ocean for an unknown period.

DSC analysis was conducted to understand the melting behavior of the compound materials and assist in determining the appropriate EBAM parameters. Figure 4 illustrates the DSC scans of both compounded filaments, revealing a single melting and crystallizing peak for each. There is no notable difference in the melting behavior between the two compounds. Based on

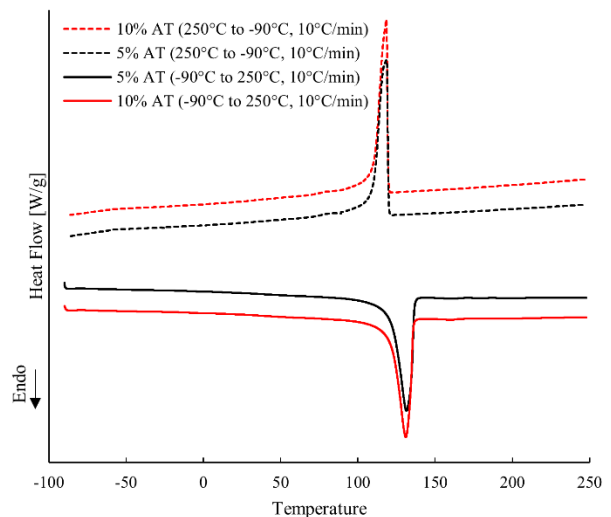


Figure 4. DSC thermograms of the compounded filaments.

the DSC results, it is recommended to set the nozzle temperature above 150°C to ensure satisfactory melting. However, the low melting point suggests that the material will soften and lose mechanical integrity at a low temperature, indicating the need for a high cooling fan speed to rapidly cool the layers and prevent excessive heating of the part.

Tensile tests were performed on both the filaments and the manufactured tensile test specimens, and a comparison was made. Figure 5 presents the comparison between the 5 and 10 wt% Acti-tech (AT) compounds, as well as between the filament and the printed part. The yield strength is lower with the higher content of the soft AT. It is observed that the yield strength remains consistent from filament to print, but the apparent E-modulus is lower for the manufactured part. It is important to note that the strength of the manufactured part was measured solely in the fibre direction of the printed path, as the interlayer strength of these compounds is generally low. This low interlayer strength becomes apparent when bending thin-walled parts by hand, as minimal force is required to separate the layers.

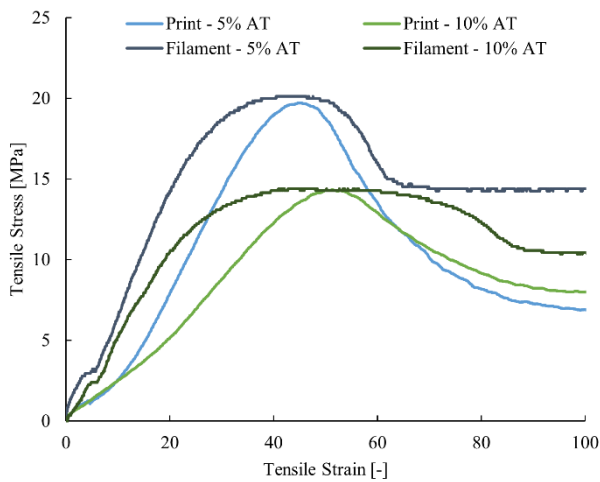


Figure 5. Tensile test performed on AM manufactured tensile bars and the filaments themselves.

Printing strategies for highly warping materials involve using high extruder temperature, slow printing speeds, and thin layers. In fact, employing low temperatures (< 250°C), high printing speeds (> 80 mm/s), and thicker layers (> 0.2 mm) resulted in rapid and complete failure of the fabrication process. Typically, minimizing the temperature difference between the extrudate and the deposited layer is achieved by heating up the build chamber. However, with this material, heating the build chamber to 40°C caused the parts to soften and fail. The degradation of the polymer material, which has been exposed to the ocean for an unknown duration, cannot be overlooked.

During filament testing, it was discovered that the nozzle temperature should be around 250°C to reduce die swell, ensure homogeneous flow, and improve layer adhesion.

To evaluate the warping of the material, a warp test part was printed according to the procedure outlined in section 2, using the parameters listed in Table 1. It was not possible to complete the test using filament with 5 wt% AT, as delamination occurred, resulting in a failed part. Therefore, only the filament containing 10 wt% AT was used to assess warping. The design of this part allowed for material contraction, and a generic PLA filament was used as a benchmark for the warp index comparison. The average warp index of the PLA part across the entire height of the sample was 4.5, exhibiting the expected behaviour of PLA.

The level of warping observed in the ocean waste material is depicted in Figure 6. Compared to generic PLA, the ocean waste exhibited two to ten times more warping, making it highly challenging to work with. The warping was so severe that it caused buckling of the part. Consistent with existing literature, reducing fabrication speed and layer thickness effectively reduced warping. Attempting to minimize the temperature difference in the build chamber by initially pre-heating it to 40°C resulted in failure, as the cooling fan was activated and caused the part to soften and fail. The material's very low melting point necessitates fast cooling, requiring the cooling fans to be set to their maximum speed. Lowering the printing temperature only led to further failure.

Based on the warping test, it is evident that these particular recycled ocean filaments cannot be viably used. The coefficient of thermal expansion (CTE) is too high, and the compatibilizer does not provide sufficient strength for interlayer adhesion. The incorporation of a stiffer compatibilizer might help minimize warping. In order to integrate these ocean plastic waste materials into an additive manufacturing life cycle, the CTE would need to be reduced through the addition of filler additives.

However, adding a filler may worsen interlayer adhesion, necessitating the use of additional compatibilizer. To achieve a sufficiently low CTE for acceptable warping, it might require a filler content of 10-30 wt% and a compatibilizer content of 5-10 wt%. In such cases, it is worth considering whether it is environmentally conscious to waste virgin materials solely to make the material viable for EBAM, or if alternative use cases for recycled ocean plastic would be more suitable. For instance, hot pressing or injection moulding may be better suited for this quality of material.

Table 1 Fabrication parameters

Part number	Extruder temp. [°C]	Build plate temp. [°C]	Fan speed [%]	Print speed [mm/s]	Layer height [mm]	Pre heat chamber to 40 °C	Print failure
01	250	25	100	30	0,2	No	At 30 mm
02	250	25	100	80	0,2	No	At 20 mm
03	250	25	100	30	0,1	No	No failure
04	250	25	100	10	0,2	No	No failure
05	250	80	0	30	0,2	Yes	Complete failure
06	250	80	100	10	0,2	Yes	No failure
07	200	25	100	10	0,2	No	Complete failure

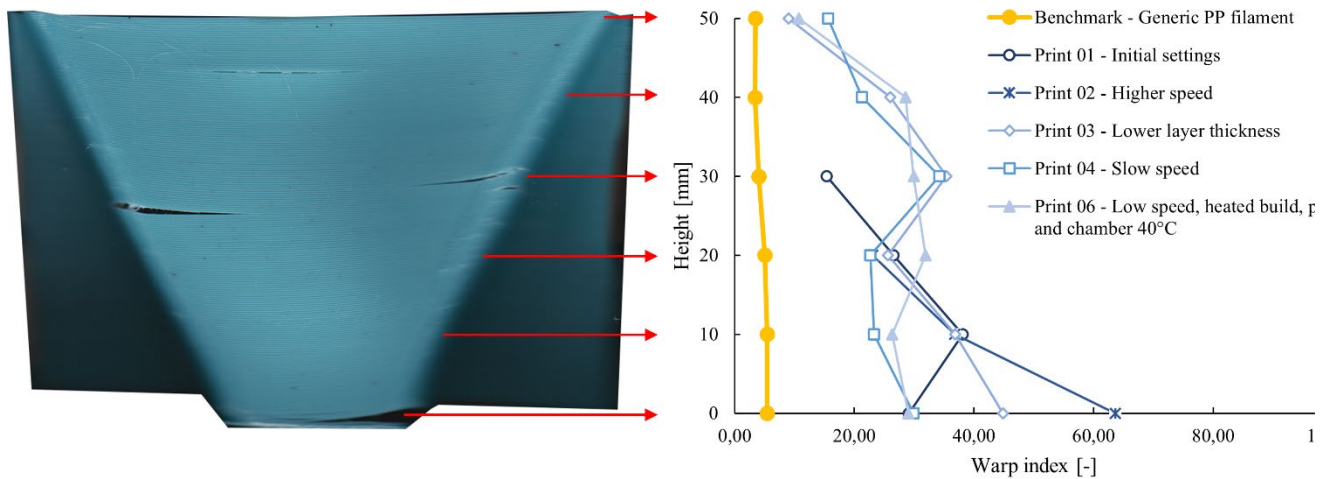


Figure 6. The printed warping part and the warp index at different heights of the fabricated samples. The part on the left is Print 03.

4. Conclusion

The degraded and unknown nature of ocean plastics presents significant challenges when working with them. In this particular study, the approach was to minimize the use of additives and only incorporate compatibilizer to improve material homogeneity. By relying solely on compatibilizers, it would theoretically facilitate easier recycling of the material afterwards. The filaments were successfully printable, but careful consideration must be given to print settings and the type of parts being fabricated. The substantial shrinkage that occurs during material cooling resulted in unintended warping of the test specimens. Therefore, while this part may not be the most suitable for evaluating high warping materials, it is valuable for optimizing print settings and minimizing warping for less problematic materials. Additionally, this part allows for scalability, enabling multiple replicas to be placed on the build plate to assess warping across the entire build volume.

Based on the experiments and results, it should be noted that using ocean plastic waste is not impossible; however, its nature renders it suboptimal for EBAM. Injection moulding or similar moulding techniques would be better suited for this material. Moreover, it is crucial to explore strategies that prevent plastics from ending up in the oceans in the first place.

References

- [1] M. S. Alsoufi and A. E. Elsayed, "Warping deformation of desktop 3D printed parts manufactured by open source fused deposition modeling (FDM) system," *Int. J. Mech. Mechatronics Eng.*, vol. 17, no. 4, pp. 7–16, 2017.
- [2] A. Armillotta, M. Bellotti, and M. Cavallaro, "Warping of FDM parts: Experimental tests and analytic model," *Robot. Comput. Integr. Manuf.*, vol. 50, no. August 2017, pp. 140–152, 2018, doi: 10.1016/j.rcim.2017.09.007.
- [3] C. McIlroy and P. D. Olmsted, "Disentanglement effects on welding behaviour of polymer melts during the fused-filament-fabrication method for additive manufacturing," *Polymer (Guildf.)*, vol. 123, pp. 376–391, 2017, doi: 10.1016/j.polymer.2017.06.051.
- [4] C. S. Davis, K. E. Hillgartner, S. H. Han, and J. E. Seppala, "Mechanical strength of welding zones produced by polymer extrusion additive manufacturing," *Addit. Manuf.*, vol. 16, pp. 162–166, 2017, doi: 10.1016/j.addma.2017.06.006.
- [5] R. Comminal, M. P. Serdeczny, D. B. Pedersen, and J. Spangenberg, "Numerical modeling of the strand deposition flow in extrusion-based additive manufacturing," *Addit. Manuf.*, vol. 20, pp. 68–76, 2018, doi: 10.1016/j.addma.2017.12.013.
- [6] E. R. Fitzharris, N. Watanabe, D. W. Rosen, and M. L. Shofner, "Effects of material properties on warpage in fused deposition modeling parts," *Int. J. Adv. Manuf. Technol.*, vol. 95, no. 5–8, pp. 2059–2070, 2018, doi: 10.1007/s00170-017-1340-8.
- [7] M. Kuzmanović, L. Delva, D. Mi, C. Martins, L. Cardon, and K. Ragaert, "Development of Crystalline Morphology and Its Relationship with Mechanical Properties of PP/PET Microfibrillar Composites Containing POE and POE-g-MA," *Polymers (Basel)*, vol. 10, no. 3, p. 291, Mar. 2018, doi: 10.3390/polym10030291.
- [8] K. Parthy, "low-bondage warp-index," 2015.