SUSTAINABLE WASTE MANAGEMENT IN ADDITIVE MANUFACTURING: OLD AND NEW STRATEGIES TO DIVERT INDUSTRIAL WASTE FROM LANDFILLS

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<u>Abstract</u>

The increasing adoption of HP Multi Jet Fusion (MJF) 3D printing in industries like orthotics has led to a surge in PA11 powder waste. Current solutions, such as in-house and third-party recycling, present challenges regarding material degradation, cost, and expertise. This paper reviews and discusses the existing strategies and proposes a novel alternative: incorporating discarded PA11 powder into bituminous binders for infrastructure construction. Preliminary findings suggest that PA11 acts as a bitumen filler, indicating this strategy's potential to divert industrial waste from landfills and replace virgin resources. This approach aligns with circular economy principles and offers a sustainable solution for managing PA11 powder waste.

Introduction

As industries increasingly adopt 3D printing to produce everything from consumer goods to medical devices, understanding its impact on the environment is becoming increasingly crucial. This becomes particularly relevant in the case of HP's Multi Jet Fusion (MJF) technology, which has been found to be applicable to the mass production of different orthotics. While other additive technologies are still in the low-volume, highvalue domain, this perfect match between MJF and its application in high-volume production of orthotics has opened the next level of challenges - what to do with its equally massive amount of industrial waste. In this paper, we'll explore and discuss some current ad-hoc solutions and propose a potentially better and more sustainable solution that can divert this industrial waste from landfills, where it will eventually end.

Multi Jet Fusion technology significantly advances additive manufacturing, combining speed, precision, and material efficiency to produce high-quality parts [1]. The process begins by applying a thin, fine-grained powder layer across the build platform. The unique aspect of MJF lies in its agent-based approach to fusing powder particles. An array of printheads deposits a fusing agent in specific areas of the powder layer where the material is intended to solidify. Simultaneously, a detailing agent is applied to define the edges of the part, enhancing resolution and surface finish. Once the agents are applied, infrared lamps pass over the build area, providing the necessary energy to fuse the powder particles in the specified regions. This combination of chemical agents and thermal energy allows for precise control over the fusing process, enabling the creation of intricate

geometries and fine details that are essential for custom and personalised devices. The process is repeated layer by layer, building the part from the bottom up until the complete object is formed [1].

In the context of orthotic production, MJF has revolutionised the production and customisation of orthotic devices, addressing many limitations of traditional manufacturing methods. The precision, speed, and material versatility of MJF enable the creation of orthotic devices tailored to individual patient needs, offering significant improvements in comfort, functionality, and overall patient outcomes. This personalisation of orthotics is one of the major advancements 3D printing in general, has provided. Using advanced 3D scanning technologies, detailed digital models of a patient's anatomy are created and then used to design devices that perfectly match the unique contours and biomechanics of the patient's body. This level of personalisation is crucial in orthotic care, where a device's fit and functionality directly impact its effectiveness and the patient's comfort. For example, in the production of custom foot insoles, 3D printing technology allows for the precise replication of the patient's foot arch and pressure points, resulting in insoles that provide optimal support and reduce the risk of discomfort or injury. Furthermore, it significantly reduces the lead time from design to final product. Traditional methods of orthotic fabrication often involve manual processes and multiple fittings, which can extend the production timeline to several weeks. In contrast, MJF technology can produce a finished orthotic device within a day. This rapid turnaround is particularly beneficial for patients requiring urgent care, such as those recovering from surgeries or injuries [2-4].

However, the main advantage MJF provides is its ability to produce parts with perfectly matching mechanical properties, which are essential for durability and performance. Although one of the main limitations of MJF is the small number of available materials, polyamide 11 (PA11) perfectly aligns the performance demand of orthotic devices and the material's mechanical properties, making it an ideal material for these applications [5]. Its high strength and durability ensure that orthotic devices can provide the necessary support and withstand the rigours of daily use. The flexibility and comfort offered by PA11 enhance patient compliance and overall effectiveness. Furthermore, the lightweight nature of PA11 makes it suitable for devices that need to be worn for extended periods, while its biocompatibility and chemical resistance ensure patient safety and device longevity.

As with any powder bed fusion processes, MJF included, powders used during printing are exposed to high ambient temperatures but only a portion of the exposed powder is fully melted and solidified. In MJF, at least 80% of the powder deposited in a manufacturing cycle often remains unmolten, and typically, only 70% of this leftover powder is collected and used for the next manufacturing cycle [6]. Consequently, roughly one-quarter of the powder per manufacturing cycle is discarded. Such a cycle incurs significant cost, logistics, and above all, environmental impact, as most of this leftover powder ends up in landfills as industrial waste.

Cost and Impact of PA11 Powder Production and 3D Printing

PA11 production begins with castor oil extraction from castor beans, primarily grown in India and Brazil. The beans are harvested and pressed to yield castor oil, which consists of approximately 85-90% ricinoleic acid. This phase is relatively energy-efficient, requiring about 3-5 GJ per ton of castor oil and producing an estimated 300-500 kg CO2e per ton due to agricultural and transportation activities [7]. Castor oil's renewable nature makes it a sustainable feedstock for PA11 production. Castor plants thrive on marginal lands unsuitable for food crops, reducing the competition for agricultural resources. The oil extraction process involves mechanical pressing and solvent extraction, with the latter being more efficient but requiring careful handling to avoid environmental contamination [8]. Once produced, castor oil undergoes hydrolysis to produce ricinoleic acid, followed by esterification and hydrogenation to form undecylenic acid, which is subsequently converted to 11-aminoundecanoic acid, the monomer for PA11. This energy-intensive stage consumes approximately 5-10 GJ per ton of monomer and emits around 500-1000 kg CO2e per ton. The chemical transformation involves multiple steps including: breaking down triglycerides in castor oil to release ricinoleic acid (hydrolysis), converting ricinoleic acid to methyl ricinoleate (esterification), heating methyl ricinoleate to produce undecylenic acid and heptanal (pyrolysis), and, reducing undecylenic acid to 11aminoundecanoic acid (hydrogenation). These processes require significant thermal and chemical energy inputs, contributing to the overall carbon footprint of PA11 production. The polymerisation of 11-aminoundecanoic acid into PA11 involves condensation reactions under controlled conditions, requiring about 3-7 GJ per ton of PA11 and producing 300-800 kg CO2e per ton. These estimates are relatively optimistic as the process is carried out in batch reactors, where temperature and pressure are controlled to achieve high molecular weight polymers, whereby the energy consumption and emissions are influenced by the efficiency of the reactors and the energy source used (e.g., natural gas, electricity) [9-10]. PA11 is then processed into powder form suitable for 3D printing through cryogenic grinding and sieving. Cryogenic grinding involves cooling the PA11 polymer to very low temperatures using liquid nitrogen, making it brittle and easier to grind into fine particles. This step requires about 2-4 GJ per ton for grinding and an additional 1-2 GJ per ton for milling and sieving, with a combined carbon footprint of 300-700 kg CO2e per ton. Overall, the total estimate for energy consumption is 15.5-30 GJ per ton of PA11 powder, with a total carbon footprint of 1550-3300 kg CO2e per ton of PA11 powder. These estimates vary significantly based on the local energy mix (renewable vs. fossil fuels) and process efficiency. Also, this estimation considers direct energy usage and process emissions but may exclude some indirect factors like equipment manufacturing [11].

When considering 3D Printing with PA11, there are some additional costs. Before printing, PA11 powder undergoes preparation, including sieving and blending, consuming approximately 1-2 kWh per kg of PA11 and producing 0.75 kg CO2e per kg. Since the MJF method involves fusing PA11 powder using a heat source, there is an energy consumption estimate of 63 kWh per kg of material with a significant associated carbon footprint of around 31.5 kg CO2e per kg, reflecting the high energy demands of the MJF process. Post-processing steps such as cooling, cleaning, and finishing add another 2-4

kWh per kg and approximately 1.5 kg CO2e per kg to the total environmental impact. Therefore, the cumulative energy consumption for producing and 3D printing 1 kg of PA11 powder using MJF technology is approximately 67.5 kWh, resulting in a total carbon footprint of 33.75 kg CO2e. These values underscore the energy-intensive nature of the process, but they also highlight the potential advantages of using a renewable resource. A typical average-sized orthotics lab producing 86,000 orthopedic insoles consumes 2.2 metric tonnes of PA11 powder per printer annually. This equates to 149 MWh of energy and 74 tonnes of CO2e. Considering that, on average, one-quarter of the powder gets discarded (0.55 tonnes of powder, 38 MWh of energy, 19 tonnes of CO2e), it clearly raises the question of how environment-friendly the process is, especially if this 0.55 tonnes of the discarded powder gets disposed to the landfill as industrial waste. Given that even a small 3D printing bureau owns more than one printer, these figures can quickly become significant.

Current Strategies of Dealing with the Discarded Powder

Currently and most commonly, the rejected powder ends up in landfills despite the existence of available alternatives. This unsustainable practice and poor alternatives may be tolerable for some time; however, more sustainable and environment-friendly strategies are needed. Currently, there are two alternatives, with their own challenges, limiting their larger uptake: in-house recycling and third-party recycling.

In-House Recycling: MJF printers are equipped with a built-in mixing chamber designed to facilitate the reuse of unfused PA11 powder from previous builds. This inhouse recycling process is a key feature promoted by HP to reduce material waste and enhance the overall efficiency of the MJF workflow. The process consists of three steps:

- 1. **Collection:** After completing a print job, the MJF printer automatically collects the unfused PA11 powder remaining in the build chamber. This powder is still usable, as it has not been exposed to the fusing agents or undergone the sintering process.
- 2. **Mixing:** The collected unfused powder is then transferred to the mixing chamber, where it is combined with 30% of the new PA11 powder. This mixing step aims to replenish the powder bed with virgin material to maintain optimal printing properties and ensure consistent part quality.
- 3. **Replenishment:** The newly mixed powder is subsequently used to replenish the powder bed for the next build. This continuous cycle of collection, mixing, and replenishment allows for the reuse of a significant portion of the unfused powder within the MJF printer.

The in-house recycling process is a valuable feature that reduces the amount of PA11 powder that would otherwise be discarded. However, the need for a refresh rate indicates that complete closed-loop recycling within the MJF printer is not yet achievable. In our previous work [2], we researched the effects of minimising the percentage of adding new powder by comparing the thermal and morphological characteristics of new, 30:70 mixed, and 100% used PA11 powder. We found that the used PA11 powder exhibited minor differences in thermal behaviour compared to the new powder, with a slight increase in melting temperature, possibly due to thermal aging. However, this did not impact

processing within MJF temperature limits. Significant differences were found in particle size distribution, where the used-PA11 powder had a narrower distribution, negatively impacting its flowability and surface fractal compared to the new powder with a wider distribution. Decreased flowability and increased surface fractal of the used powder led to rougher build layer surfaces, resulting in higher porosity in fabricated specimens, primarily between layers. As shown in the figure below, this increased porosity directly correlates with inferior mechanical performance, particularly in tensile strength, compared to specimens made with new and 30:70 mixed PA11 powder.



Figure 1: Tensile stress-strain curves for specimens fabricated with new, 30:70 mixed, and used PA11 powder (reproduced from Pandelidi *et al.*[6]).

Third-Party Recycling: Several third-party companies have emerged, offering solutions for recycling PA11 powder that has exceeded its recommended reuse within MJF printers. One such company is Nexa3D. Their Quantum Laser Sintering (QLS) technology uses the rejected PA11 powder that has exceeded its recommended lifespan in MJF printers and has shown promising results in enhancing the recyclability of PA11 powder [12-14]. QLS is a proprietary laser sintering technology developed by Nexa3D that utilises a high-power CO2 laser to fuse powdered materials. The technology boasts faster printing speeds and improved energy efficiency than traditional selective laser sintering (SLS) methods. QLS is particularly well-suited for recycling PA11 powder from MJF printers due to its ability to process materials with varying properties and its compatibility with a wide range of powder bed fusion parameters. Implementing QLS for PA11 powder recycling involves several steps:

1. **Powder Collection:** The used PA11 powder is collected from MJF printers after reaching its maximum recommended reuse cycles.

- 2. **Powder Preparation:** The collected powder may undergo pre-processing steps, such as sieving or drying, to ensure optimal quality and consistency for QLS processing.
- 3. **QLS Printing:** The prepared PA11 powder is then loaded into a Nexa3D QLS printer and processed according to the specific parameters required for the desired part or application.

While QLS offers a promising solution for PA11 powder recycling, the recycled PA11 powder exhibits variations in properties compared to virgin material, and as discussed earlier, impacts the mechanical performance of printed parts [2], and in certain demanding applications, may not be suitable at all. Nexa3D deals with this by relying on their design expertise, particularly by assessing and applying suitable Factors of Safety [12]. A lack of this expertise, especially when dealing with demanding applications, may pose barriers to adopting this strategy.

Arkema's Virtucycle Program: This is a comprehensive initiative by Arkema, the supplier of the PA11 powder, aimed at promoting the circularity and sustainability of their high-performance polymers, including PA11 and PA12 [15-17]. The Virtucycle Program focuses on establishing closed-loop and open-loop recycling circuits for Arkema's polymers. It involves the collection of post-industrial and post-consumer waste, which is then processed and transformed into high-quality recycled materials. The Virtucycle Program operates on a multi-pronged approach, encompassing the following key stages:

- 1. **Collection and Sorting:** Arkema collaborates with a network of partners to collect post-industrial and post-consumer waste containing their polymers. The collected waste undergoes meticulous sorting to ensure the quality and purity of the recycled material.
- 2. **Recycling and Regeneration:** The sorted waste is then processed at Arkema's state-of-the-art recycling facilities. This involves depolymerization, purification, and repolymerization processes, transforming the waste into high-quality recycled polymers.
- 3. Certification and Traceability: Arkema subjects the recycled polymers to rigorous quality control measures and provides certifications to ensure their compliance with industry standards. The company also maintains a robust traceability system to track the origin and journey of the recycled materials.
- 4. **Marketing and Distribution:** The certified recycled polymers are marketed under the Virtucycle brand, offering customers a sustainable alternative to virgin polymers with comparable performance characteristics.

The program offers various levels of participation to subscribing companies, allowing companies to choose the most suitable option based on their needs and capabilities, and it has garnered significant acclaim for its success in diverting plastic waste from landfills and reducing the environmental footprint of high-performance polymers. However, despite its notable successes, the Virtucycle Program faces certain challenges. Collecting and sorting post-consumer waste can be complex and costly, particularly in regions with inadequate waste management infrastructure. Additionally, the depolymerisation and repolymerisation processes require significant energy inputs, raising concerns about the overall environmental impact. Furthermore, the economic viability of recycled polymers may vary depending on market conditions and the specific applications.

A New and Proposed Alternative Strategy

An alternative approach to the strategies discussed above is to incorporate the powder in bituminous binders, as this tends to be a common practice with recycled polymers. In general, infrastructure construction has been identified as one of the sectors with the capacity to absorb significant amounts of waste and divert it from landfills [18]. For example, the incorporation of polymers, including polypropylene (PP) [19, 20], highdensity polyethylene (HDPE) [20, 21], low-density polyethylene (LDPE) [20-22], and acrylonitrile-butadiene-styrene (ABS) [21-23], in bitumen were investigated. Overall, their addition has resulted in an increase in viscosity [16], an increase in softening point, and a decrease in penetration values [19, 21, 23-25]. However, one of the barriers to adopting such materials was the presence of additives and contaminants on their surface, which deteriorates the performance of resulting products [19]. It is strongly highlighted that new waste materials should only be accepted if they consistently and reliably do not adversely affect current practices. Industrial waste, like the MJF's PA11, is expected to be free of such contaminants, creating an opportunity to solve the recycling problems by developing innovative materials in infrastructure construction. However, unlike other polymers such as polyethylene (PE), PP, polyvinyl chloride (PVC), polystyrene (PS), polyethylene terephthalate (PET), and ABS, which were extensively studied in the literature [27, 28], research on the use of polyamides has been very limited.



Figure 2: FTIR spectra for C170, PA11, C170/3.5 wt.% PA11, C170/7 wt.% PA11, and C170/10 wt.% PA11

Therefore, in this preliminary work, an investigation into the possibility of using PA11 as an additive to bitumen was performed. In particular, the work was focused on understanding the interactions between PA11 and C170 bitumen following blending. On three different blends in 3.5, 7, and 10 wt% concentrations, Fourier-transform infrared

(FTIR) spectroscopy was undertaken to investigate whether any chemical interaction took place during blending. FTIR was conducted in the infrared region of 4000-600 cm-1 using a Perkin-Elmer spectrum 2 device in ATR mode. Each spectrum was acquired on 32 scans at 4 cm-1 resolution and a scan speed of 0.2 cm/s.

The figure above presents the preliminary results - the FTIR spectroscopy spectra for C170, PA11, and all blends. The peaks at 2920 and 2850 cm-1 for asymmetric and symmetric stretching of CH2, respectively, were found for both the PA11 and C170. For PA11, the following were observed:

- 1. the absorbance at 3306 cm⁻¹ reflects N-H stretching,
- 2. the absorbance at 1634 cm^{-1} is for the C=O stretching of Amide I,
- 3. C-N stretching and C=O in-plane bending are detected by the peak at 1537 cm⁻¹,
- 4. the absorbance at 1470 cm⁻¹ can be attributed to C=O and N-vicinal CH2 bending,
- 5. the absorbance peak at 1223 cm^{-1} is a result of C-N stretching, and
- 6. that at 683 cm^{-1} is for CONH out-of-plane deformation of Amide V [25].

The C170 presents an absorbance peak at 1600 cm⁻¹, attributed to the presence of aromatic rings and two peaks due to aliphatic groups -CH3 and -CH2 around 1375 and 1460 cm⁻¹, respectively [29]. The binders presented peaks consistent with those of C170, and no new peaks could be identified. This indicated that new chemical groups were not created following the blending process, or at least to no significant extent to be detected. FTIR, when performed at ATR mode, has a penetration depth of approximately 3 μ m depending on the experimental conditions [30], so, although PA11 particles were certainly present on the crystal, they were coated by bitumen well enough so that the PA11 characteristic peaks were not detected by the binder samples.

These findings suggest that the interactions between the C170 and PA11 are most likely purely physical, suggesting that the PA11 acts as a filler in the bitumen rather than a modifier. This is the most favourable result and suggests a great potential for this strategy, as it not only diverts the industrial waste from the landfill but also has the potential to fully replace the virgin resource currently used as the filler.

Conclusion

The increasing adoption of HP Multi Jet Fusion (MJF) 3D printing in various industries, notably orthotics, has amplified concerns about the environmental impact of PA11 powder waste. Current strategies to address this issue include in-house recycling by reusing the powder in MJF printers and third-party recycling services like Nexa3D's Quantum Laser Sintering technology. While in-house recycling offers convenience, it is limited by material degradation over multiple reuse cycles. Though more comprehensive, third-party recycling can be costly and require specialised expertise. Arkema's Virtucycle Program offers a closed-loop solution by collecting and regenerating post-industrial and post-consumer PA11 waste, but it faces challenges related to collection infrastructure and energy-intensive processes associated with repurposing the powder. A new alternative is proposed and incorporates discarded PA11 powder into bituminous binders for road construction, a strategy supported by preliminary findings suggesting PA11 acts as a filler

in bitumen. This approach aligns with circular economy principles, offering a potentially sustainable and economically viable solution for managing PA11 powder waste while contributing to infrastructure development. While technologically viable, further research is needed to validate this strategy's long-term performance and environmental impact.

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