Journal of
Composites Science

MDPI

Review

Mechanical Performance of Recycled 3D Printed Sustainable Polymer-Based Composites: A Literature Review

Ioannis Filippos Kyriakidis [1,2], Nikolaos Kladovasilakis [2], Eleftheria Maria Pechlivani [2] and Konstantinos Tsongas [1,*]

[1] Advanced Materials and Manufacturing Technologies Laboratory, Department of Industrial Engineering and Management, School of Engineering, International Hellenic University, 57001 Thessaloniki, Greece; giankyri@iem.ihu.gr
[2] Information Technologies Institute, Centre for Research and Technology Hellas, 57001 Thessaloniki, Greece
* Correspondence: ktsongas@ihu.gr

Abstract: The development of efficient waste valorization strategies has emerged as an important field in the overall efforts for alignment with the environmental goals that have been set by the European Union (EU) Green Deal regarding the development of sustainable circular economy models. Additive manufacturing has emerged as a sustainable method for secondary life product development with the main advantages of it being a form of net-zero waste production and having the ability to successfully transport complex design to actual products finding applications in the industry for rapid prototyping or for tailored products. The insertion of eco-friendly sustainable materials in these processes can lead to significant reduction in material footprints and lower energy demands for the manufacturing process, helping achieve Sustainable Development Goal 12 (SDG12) set by the EU for responsible production and consumption. The aim of this comprehensive review is to state the existing progress regarding the incorporation of sustainable polymeric composite materials in additive manufacturing (AM) processes and identify possible gaps for further research. In this context, a comprehensive presentation of the reacquired materials coming from urban and industrial waste valorization processes and that are used to produce sustainable composites is made. Then, an assessment of the printability and the mechanical response of the constructed composites is made, by taking into consideration some key thermal, rheological and mechanical properties (e.g., viscosity, melting and degradation temperature, tensile and impact strength). Finally, existing life cycle analysis results are presented regarding overall energy demands and environmental footprint during the waste-to-feedstock and the manufacturing processes. A lack of scientific research was observed, regarding the manifestation of novel evaluation techniques such as dynamic mechanical analysis and impact testing. Assessing the dynamic response is vital for evaluating whether these types of composites are adequate for upscaling and use in real life applications.

Keywords: recycled materials; composites; additive manufacturing; LCA; mechanical properties; dynamic mechanical analysis

Citation: Kyriakidis, I.F.; Kladovasilakis, N.; Pechlivani, E.M.; Tsongas, K. Mechanical Performance of Recycled 3D Printed Sustainable Polymer-Based Composites: A Literature Review. *J. Compos. Sci.* **2024**, *8*, 215. https://doi.org/10.3390/jcs8060215

Academic Editor: Francesco Tornabene

Received: 15 May 2024
Revised: 30 May 2024
Accepted: 5 June 2024
Published: 7 June 2024

1. Introduction

Waste management and valorization of end-of-life materials are important issues that need to be addressed. The recycling and upcycling of wastes have emerged as a promising step towards optimized production processes and resource management, contributing to the adaptation of a circular economy as a sustainable long-term solution. Plastic waste is one of the biggest concerns that the planet is facing. Most polymers are non-biodegradable, with an estimated degradation time of 10 to 450 years. They are mainly categorized as thermoplastics or thermosets. Thermosets, although they present superior strength, once hardened, cannot be thermally processed and reshaped, meaning they are not recyclable. Thermoplastics, on the other hand, are appropriate for recycling due to their ability to withstand multiple thermal processes, making them the most suitable candidate for sustainable manufacturing [1]. The rate of recycled plastics is relatively low (9%), with approximately 85% of produced polymeric waste ending up in landfills, with the amount of generated plastic waste estimated to triple by 2060 [2,3]. The European Union (EU) adopted a new circular economy action plan (CEAP) in 2020, as a prerequisite to achieve the EU's 2050 climate neutrality target and to halt biodiversity loss. This plan contains new plastic waste recycling regulations to achieve the EU agenda of clean water and sanitation and the efficient use of marine resources and oceans to achieve the milestones of Sustainable Development Goal 6 (SDG6) and 7 (SDG7) for clean water and clean energy production. The development of sustainable water and waste management patterns from SDG6 and SDG7 is also relevant to Sustainable Development Goal 12 (SDG12) of the EU agenda for the development of responsible and sustainable consumption and production patterns to assess the rapidly growing material environmental footprint that was measured at 92 billion metric tons in 2017 and is projected to reach 192 billion metric tons by the year 2060 [4,5].

Additive manufacturing (AM) is a swiftly rising manufacturing technique with the ability to produce complex designs with high detail accuracy that allows for design freedom and the chance for near zero-waste production contrary to other conventional manufacturing processes due to its minimized material usage resulting in reduction of the material footprint [6–10]. Although plastics are the main type of materials that are utilized by AM processes, in recent years other raw materials such as metals and ceramics have also been introduced into these manufacturing processes. Metallic materials are mainly in the market in the form of alloys with titanium, aluminum and nickel-based alloys being the most known. Nickel-based alloys present exceptional mechanical properties, especially in high temperatures, making them great candidates for applications in the aerospace industry such as combustion chambers of gas turbines and for turbochargers or exhaust valves in the automotive industry. Aluminum alloys are mainly used to reduce weight in applications and have been utilized mainly for exterior design in the automotive and aerospace industry (wings, panels). Titanium alloys are mainly used for tools in the medical and dental industry, while cobalt–chromium (CoCr) alloys are identical for medical implants. Metals are mainly used in powder form in power bed fusion (PBF), wire arc direct energy deposition (DED), and binder jetting AM processes [11–16]. Ceramic materials have been used in AM processes, especially for material extrusion filament-based processes. Dut to frequent air entrapment and nozzle clogging, ceramics are not typically used as sole materials but more as powder fillers in composites. In PBF processes, the main limitations are rapid heating and cooling of the printing surface. Ceramics are unusable for applications in extremely low or high temperatures due to their brittle behavior. Another problem that occurs is the segregation of ceramic powders in VAT photopolymerization processes such as stereolithography (SLA) [17–21].

It is commonly accepted in the literature that recycled materials often exhibit significantly low mechanical properties compared to their virgin counterparts. To solve this issue, composite materials have been introduced to AM industry. Fiber-reinforced polymers provide exceptional strength-to-weight ratio finding applications in the automotive, aerospace and construction industries. Synthetic fibers are used to produce lightweight efficient materials to replace the originals. In recent years, a demand for recyclability and sustainability has been raised, with natural fibers being introduced and adopted to replace non-recyclable synthetic ones. Sustainable complex materials made from biodegradable or reused polymers with recycled reinforced materials could lead to further reduction of material footprint, energy demands and carbon emissions, leading to cleaner production processes, aligning industrial manufacturing with the milestones set from SDG12 of EU's agenda [22–26]. On this note, Rashid and Koc [1] reviewed the guidelines and circular economy models regarding the sustainability of polymer-based additive manufacturing. Parandoush and Lin [9] reviewed the printability and mechanical response of polymer-based fiber reinforced composites for material extrusion (MEX), PBF and VAT photopolymerization additive manufacturing. Lodha et al. [6] focused on the valorization of recycled materials for AM, analyzing the recycling processes especially for recycled polymers coming from urban waste and transformed to resins. Ghabezi et al. [27] focused on the valorization of industrial waste polypropylene combined with recycled carbon fibers for eco-friendly MEX AM processes, while Sealy [28] stated the importance of repurposing waste and exploitation of it for the production of sustainable polymeric composites to achieve less raw material demand and lower the environmental footprint. Although significant research regarding the possibility of utilizing AM processes for waste valorization has been conducted, limited research has been carried out regarding the exploitation of metals and ceramics on AM, especially as fillers on polymer-based recycled matrices. Also, there has not been a comprehensive analysis on the effect of the reinforcements on the mechanical response, compared to the conventional or sole recycled materials.

The aim of this specific review is to state the existing progress in additive manufacturing processes emphasizing on recycled materials and the recycling process from waste to feedstock for AM, especially feedstock coming from industrial waste, and reviewing the mechanical response of the proposed sustainable polymer-based composite materials, to assess the issues that have been stated according to SDG12. In Section 2, the printability of those feedstocks is stated, taking into consideration their thermal and rheological properties in combination with the optimization of the printing conditions. Section 3 introduces crucial mechanical properties and static analysis methods for their quantification. It emphasizes the importance of innovative methods such as impact and dynamic mechanical analysis (DMA) for a more comprehensive analysis, and it states the urgency for the evaluation of the final composites recyclability through life cycle assessment (LCA) analysis. In Section 4, possible applications of the structured composites are investigated taking into consideration large-scale prototypes. A brief presentation of the sections contained in this specific review is presented in Figure 1, while Table 1 lists the acronyms used throughout this report.

Table 1. Table of acronyms.

Meaning	Acronym	Meaning	Acronym
Additive Manufacturing	AM	Selective Laser Melting	SLM
Injection Molding	IM	Selective Laser Sintering	SLS
Power Bed Fusion	PBF	Laser Metal Deposition	LMD
Direct Energy Deposition	DED	Liquid Deposition Modeling	LDM
Stereolithography	SLA	Gas Atomization	GA
Dynamic Mechanical Analysis	DMA	Plasma Rotating Electrode Process	PREP
Recycled	r	Melt Flow Index	MFI
Polyethylene Terephthalate	PET	Weight Fractions	wt.%
Polyethylene Terephthalate Glycol	PETG	Pyromellitic Dianhydride	PMDA
High-Density Polyethylene	HDPE	Ethylene–Ethyl–Acrylate	EEA
Low-Density Polyethylene	LDPE	Multi-Walled Carbon Nanotubes	MWCNTs
Polyvinyl Chloride	PVC	Carbon Fibers, Carbon Black, Carbon Nanotubes	CF, CB, CNTs
Polypropylene	PP	Tamarind Fruit Shells	TFS
Polystyrene	PS	Corn Husk Fibers	CHF
Acrylonitrile Butadiene Styrene	ABS	Cocoa Beans Shells	CBS
Ethylene–Vinyl Acetate	EVA	Thermoplastic Polyolefins	TPOs
Ground Tire Rubber	GTR	Polyethylene-Grafted Maleic Anhydride	PE-g-MA
Polycarbonate	PC	Young's Modulus	E
Polylactic Acid	PLA	Diffuse Light Stereoscopy	DLS
Thermoplastic Polyurethane	TPU	Thermogravimetric Analysis	TGA
Material Extrusion	MEX	Digital Image Correlation	DIC
Fused Filament Fabrication	FFF	Finite Element Analysis	FEA
Fused Granular Fabrication	FGF	Ultimate Tensile Strength	UTS
Fused Particle Fabrication	FPF	Global Warming Potential	GWP
Fused Deposition Modeling	FDM	Cumulative Energy Demand	CED
Electron Beam Melting	EBM	Life Cycle Assessment	LCA

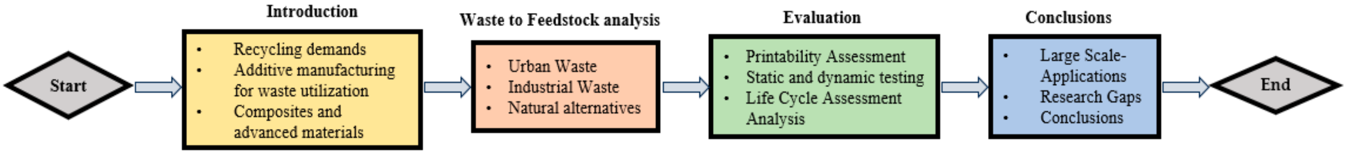


Figure 1. Flowchart of the review paper.

2. Recycled Materials and the Introduction of Them in Additive Manufacturing

2.1. Plastics, Polymeric Blends and Polymeric Composites as 3D Printing Raw Material

In recent years, a plethora of studies have been focused on the valorization of waste or reused polymeric materials for AM, including the use of conventional recycled biodegradable polymers, with 7 types of recycling plastics identified to date. Different types of polyethylenes have been utilized for the production of high-volume plastic products such as bottles and packaging, replacing conventional polyvinyl chloride. Stronger thermoplastics such as polypropylene and styrene-based polymers have been used for automotive or aerospace applications. Flexible polymers such as polylactic acid or thermoplastic polyurethane have been utilized for everyday equipment [29–31]. Table 2 presents an overview of the types of plastics that have been researched for recycling, assessing recyclability and possible future applications. Regarding the recycling source and the state of materials, recyclable polymers or polymeric mixtures (blends) can be acquired.

Table 2. Brief presentation of the plastics collected for recycling.

Recycling Symbol	Plastic (Acronym)	Recyclability	Applications
♳ PET	Polyethylene Terephthalate (PET)	Easy	Packaging, textiles, electronic cables, water bottles [32–40]
♴ HDPE	High-Density Polyethylene (HDPE)	Easy	Pipes, chemicals packaging, fuel tanks, bumpers [41–46]
♵ PVC	Polyvinyl Chloride (PVC)	Moderate	Bags, medical equipment, pipes [47]
♶ LDPE	Low-Density Polyethylene (LDPE)	Moderate	Bags, containers, sporting equipment [48–50]
♷ PP	Polypropylene (PP)	Difficult	Automotive components, turbine blades, medical tools [51–58]
♸ PS	Polystyrene (PS)	Difficult	Insulation products, disposable cups [59–65]
♹ OTHER	Acrylic (ABS), Polycarbonate (PC), Polyamide (PA), Thermoplastic Polyurethane (TPU), etc.	Difficult	Wheel covers, bumpers, headlight lenses, safety equipment [59–65]

2.1.1. Urban Polymeric Waste

PET is usually used in AM by employing material extrusion (MEX) processes such as fused filament fabrication (FFF), but several challenges have emerged due to its shrinkage and warpage issues caused by high fusion temperatures and lack of control of crystallinity, water absorption (leading to molecular weight reduction) and weak interfacial bonding between layers [32–35]. PET is mainly used for water and food packaging and the waste-to-feedstock process consists of label removal, water cleaning and drying, and granulation or shredding into flakes or pellets before sterilization. Single- or twin-screw extruders are commonly used for feedstock production [36–40]. Polyethylenes (HDPE and LDPE) are used as well in the same industry, with HDPE providing a high strength-to-density ratio and being employed for blow-molded water bottles. In contrast to LDPE, HDPE has significantly higher tensile strength and higher intermolecular force due to the absence of branching [41,42]. The exploitation of reused or recycled HDPE could offer long-term benefits. In AM, frequent use of HDPE composites in pelletized form is used for MEX 3D printing by following the same steps for the waste-to-filament process as those described for PET [43,44]. Another source for reliable acquisition of recycled HDPE or PET could be plastic waste that ends up in the natural environment (oceans, rivers, etc.), which is mainly found in the form of microplastics. The acquisition of this waste is harder in these environments in comparison to landfill waste because of the degradation of the polymer in those circumstances and other factors, but the status of the collected waste could make the waste-to-filament procedure simpler [45,46]. LDPE's use in AM is limited due to its poor adhesion and high shrinkage. Usually in granular form, it has been mixed with certain materials such as ceramic powders for fuse granular fabrication (FGF) AM processes [48–50]. PVC presents low printability due to the degradation of its crystalline structure at relatively low temperatures and its poor thermal stability. Recently, some efforts have been made with the combination of PVC and polylactic acid (PLA) to produce a printable feedstock for MEX AM with tailored flexibility and ductility [47].

2.1.2. Industrial Polymeric Waste

Industrial waste utilized for recycling processes mainly comes from automotive and e-waste plastics. Sterilizing is unnecessary in contrast to urban waste processing, and the quality of the collectable materials is higher due to lower contamination levels. Polypropylene is among the most frequently used thermoplastics, with applications in high-volume manufacturing such as yogurt cases, bags or packages, and more recently it has been inserted into renewable energy, with ÉireComposites™ in Ireland producing glass fiber-polypropylene wind turbine blades (GF-PP). GF/PP is an extremely tough, lightweight composite and provides for quiet and durable production of electricity from the wind, while simultaneously being recyclable [51]. However the main source of polypropylene waste is automotive industry wastes. In the UK, it has been also found in electronic wastes, while during the COVID-19 pandemic it was used to produce protective masks. Health industry wastes need to be sterilized before they can be recycled such as common urban wastes [52–55]. PP is mainly used as a single-end plastic, studies have shown that tensile properties have a steady behavior despite the common perception of plastic downgrading that has been verified for other types [56,57]. Recycled PP has shown property stability during recycling processes making it, despite the warping issues, a promising material for future investigation and valorization [53,58]. The amount of electronic waste has been rapidly increasing in the last decade, with ABS being the main polymer that can be extracted from it. ABS is one of the most suitable materials used as feedstock for 3D printing. The process from waste to feedstock for electronic wastes consists of the steps of collecting e-waste, disassembling, size reduction by shredding it in granule form and extrusion in pellet or filament extruder depending on the desired feedstock. Gaikwad et al. [60] recovered ABS and PC from end-of-life printers and created filament out of it with a slight decrease (10–20%) in breaking and tensile strength while reducing the CO_2 emissions by 28% during the AM process in comparison to the equivalent virgin materials [59,61]. Tires are recycled mainly with mechanical and cryogenic processes, for downgrading and producing 2nd life cycle products. Ground tire rubber (GTR) can be a reliable source of sustainable acquisition of thermoplastic and thermoelastic polymers. Due to the organics contained in the tires, there is a potential for them to serve as an end-life energy resource. Besides the organic load, some mentionable thermoplastic polymers that have been acquired from exploitation of ground tire rubber are PP or PC and the thermoplastic elastomer ABS [62–64]. Another thermoplastic elastomer that is appropriate for AM is TPU, which can be recovered from polyurethane foams or from 3D printing waste and can be processed to either filament form for MEX AM or in powder form for selective laser sintering (SLS) AM working as matrix material or as filler material to improve elasticity [65].

2.1.3. Natural Polymers as an Alternative Solution (PLA)

Integrating natural polymers into AM processes could be a step towards sustainability. PLA is mainly made from sugarcane or corn starches. Although it has low mechanical properties, it is considered one of the easiest printable materials to work with, making it an ideal candidate for replacing other high-volume manufacturing polymers [66]. It is easily recyclable, and blends of virgin and recycled PLA have already been made without a significant drop in mechanical properties [67,68]. For the development of stronger novel materials, nickel powders or other metals have been combined with it [69].

2.1.4. Overview of the Recycling Process of Polymers

In general, conventional polymers which are intended for recycling exhibit low degradability, and the development of an appropriate treating process is indispensable. There are many factors that are taken into consideration to define whether a plastic is easily recyclable or not. Those are its ease of collection and sorting, its main thermal properties, such as melting and degradation temperature, that affect the amount of energy demands for the treatment, and its weight loss during the continuous thermal and mechanical processing affecting economic efficiency. The aforementioned properties also affect the feedstock production process in AM, since there are stages of sudden temperature changes (heating on the nozzle on MEX AM, cooling on the bed) that can affect a material's structure and stability. Recycled plastics are mainly utilized for MEX AM techniques in filament or pellet form, or for selective laser sintering (SLS) for PBF AM in powder form. Recycled plastics in powder form are mainly used as fillers in conventional materials [70,71]. In Figure 2, a flowchart of the waste to feedstock process regarding the type of waste and the form of the exported 3D printing feedstock is presented, and the state of the art regarding the type of polymer acquired from each type of waste is presented in Table 3.

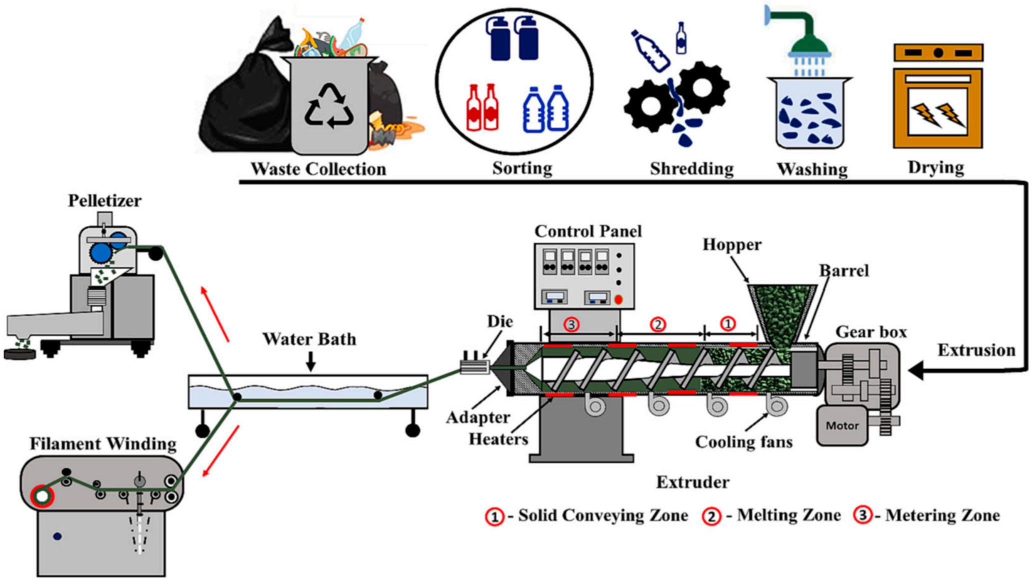


Figure 2. A general flowchart of the waste-to-feedstock process for plastic waste [70].

Table 3. Waste type and feedstock material for additive manufacturing.

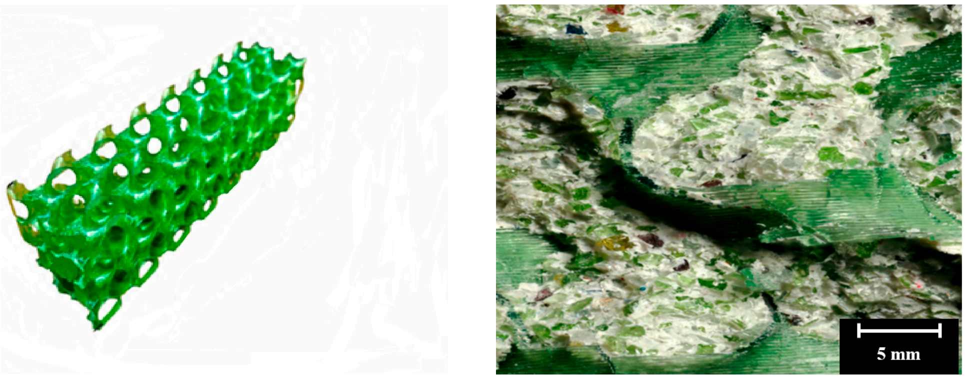
	Waste Type and Sources	Recycled Polymer	References
Urban	Plastic Bottles	PET, HDPE LDPE	[34,36,44]
	Food Packaging	HDPE, LDPE	[43,49]
	Plastic Bags	PVC, nylon	[47]
	Marine and Ocean	PET, HDPE	[44,46]
	Everyday Equipment	PP, ABS, PLA, TPU	[52]
Industrial	Automotive Parts	PP	[54]
	Ground Tire Rubber	ABS, PP	[62,63]
	Electronic Devices	ABS, PC, PS	[53,59,60]

2.2. Metal Alloys or Metallic Composites

Metals are mainly used in the aerospace, automotive, construction and medical industries. Metals that have been utilized in AM processes are mainly nickel, aluminum and titanium-based alloys. Cobalt–chromium and copper alloys have been recently introduced to additive manufacturing and have been utilized mainly with wire arc DED AM. Metals exhibit excellent mechanical strength, making them suitable for durable applications. They are mainly used in powder for PBF AM processes such as EBM and SLM or stereolithography (SLA) [72]. The effect of powder reuse has been studied for nickel-based alloy 718 [73,74] and for the titanium alloy known as ASTM grade 5 [75,76], showing no effects of the powder reuse on mechanical properties compared to the equivalent virgin metals, allowing for recycling and material cost limitation for multiple circles. Powders for metal AM are utilized mainly by gas atomization (GA), although recently the plasma rotating electrode process (PREP) or plasma atomization has been introduced. Studies have shown that PREP powders with less porosity and dilution meaning better printing quality in comparison to GA powders. Also, PREP powders had lower Nb segregation and lower Laves phase fraction resulting in better overall mechanical properties [77–79].

2.3. Ceramics

The introduction of ceramics in AM has encountered numerous challenges regarding the accuracy and actual durability of AM-constructed ceramics in comparison to other conventional manufacturing processes. They are used as matrix or fillers in composites (carbon, glass fibers, carbon nanotubes), and due to their high strength, they are suitable for structural applications. Ceramics in combination with high-volume manufacturing polymers, such as PET, as scaffolds (Figure 3) have been studied with promising results for the development of new architected materials and the incorporation of the reuse of raw materials. The addition of PET scaffolding in ceramic matrix has led to the improvement of thermal properties. Besides the drop in mechanical properties, the proposed composite could work as an example of efficient utilization of waste ceramics with additive manufacturing for achieving tailored properties for specific applications [80–82]. Concrete composites show promise due to their negative Poisson ratio resulting in great levels of energy absorption, high strength and limited shrinkage. Cementitious composites are used for high-durability applications in civil engineering, but the development of high cement/aggregate materials is resulting in higher demand for Portland-based cement, meaning higher cost and higher environmental impact [83–85].



(**a**) (**b**)

Figure 3. Cement brick composite with internal PET scaffolding: (**a**) Gyroid additive manufactured PET scaffold. (**b**) Optical microscopy image of the fracture surface of the tested composite [82].

3. Evaluation of Recycled Materials Regarding Additive Manufacturing Processes

3.1. Developing Printable Feedstocks, Material Characterization and Optimizing Printing Conditions

The quality of the printing procedure has an immediate effect on the performance of the final structure. It is important to use adequate materials for AM and assess the appropriate printing conditions for each case to exploit recycled materials in an optimal way. Physicochemical and rheological property assessment for the used materials is vital to evaluate their printability. The introduction of appropriate fillers as stabilizers or chain extenders has been shown to help improve thermal stability, an important factor due to the temperature differences between the extrusion nozzle and the printing bed. Identifying viscosity, crystallinity and melting is necessary for assessing the optimal printing conditions for efficient results.

3.1.1. Additive Manufacturing of Polymers

Composite materials have shown promise in AM but also have raised the need for optimization of the printing process. Printing conditions and demands such as specific flow rate, dimensional and schematic accuracy, and thermal and dimensional stability of the final structure are vital to secure the optimal mechanical properties and expand the range of applications AM materials can have [86]. Thermal analysis has focused on identifying the melting point and the optimal printhead temperature settings for filament granular or pellet extrusion techniques [32,33,36]. Rheological properties, such as viscosity, can have an impact on flowability mainly measured by the melt flow index (MFI), and the crystallinity of the material can lead to clogging and breakage, affecting printability [87]. Recycled PET has been utilized for AM, but due to shrinkage, weak interfacial adhesion and warping, the insertion of stabilizers and reinforcements is vital for sustainable 3D printing [45]. The addition of pyromellitic dianhydride (PMDA) or other chain extenders helps improve viscosity [35,37,88]. Recycled carbons (fibers, biochar) used as reinforcement have been shown to help limit shrinkage issues and improve thermal stability and dimensional accuracy [38,89]. In Table 4, the main printability parameters regarding PET and rPET composites are demonstrated along with the corresponding reference.

Table 4. Research assessing printability of rPET and rPET composites.

Matrix Polymer	Fillers	Weight Fractions	Printing Method	Main Findings	Reference
rPET	-	-	FPF	Particle shape and influences printability and mechanical response	[32]
			FDM	Different grades of rPET require different printing conditions	[35]
				Moisture negatively influences printing quality and mechanical properties	[36]
				Thermal stability decreases after each recycling cycle	[45]
			FFF	Fan cooling and printing bed temperature affect the crystallinity	[86]
	PMDA	0.3–0.75 wt.%	FDM	Printable filament, brittle behavior, need for further research	[37,88]
	rCFs	0.4–40.7 wt.%	FFF	Decreased shrinkage	[38]
	Biochar	0.5–5 wt.%		Increased degree of crystallinity led to better dimensional and thermal stability	[88]

Recycled HDPE is difficult to print due to warping issues and poor flow because of the high degree of crystallinity. The addition of recycled carbon fibers (rCFs) had a significant effect on the degree of crystallinity, with a drop from 63.3% to 48.7% [42], but still higher than the crystallinity of other materials such as rPET (maximum 36.3%) [87]. Kumar et al. inserted multi-walled carbon nanotubes (MWCNTs) at a wt.% from 0–5% and found a significant drop in MFI and an increase in melting point and degree of crystallinity in comparison to plain HDPE. This thermal stability, although it makes the printing process more difficult, has an immediate positive effect on the flexural modulus and strength of the structured composite [90]. The same behavior was observed through three recycling cycles of similar composites [46]. Adding bio-carbons has shown to be a possible way to solve warpage issues on rHDPE/rPP blends with a significant lowering of the composite's viscosity and increase in MFI providing improved printability for FFF AM processes [44]. Natural fibers (hemp) reinforced rHDPE's lower degree of crystallinity without raising the melting point, while the addition of polyethylene-grafted maleic anhydride (PE-g-MA) to the composite increased the crystallinity. An optimal concentration of both could be a way to produce advanced materials with tailored properties [91]. The addition of sawdust to rHDPE along with coupling agent Dupont Fusabond® E265 for better particle adhesion (45%, 45%, 10%) has been shown to help improve dimensional accuracy, an important factor for sustainable and quality large-scale AM [92]. On the other hand, rLDPE, although widely available, shows poor adhesion and high shrinkage and is one of the most challenging materials to be used as a matrix for AM with an observed crystallization peak at 90 °C, highlighting the importance of further research on rheological properties [50]. Oberloier et al. [93] tested the efficiency of the particle swamp optimization (PSO) experimenter for the identification of the optimal printing parameters of rLDPE which could be exploited for printing optimization of unknown or before unprintable materials. High-strength fillers such as SiC/Al_2O_3 nanocomposites or CNTs inserted into rLDPE have shown potential to improve thermal stability and wear factor over repeated heating and cooling processes [48] and overall better MFI [49,94], resulting in overall better rheological and mechanical properties in comparison to virgin LDPE. An overview of the conducted research regarding recycled polyethylenes is stated in Table 5. Another challenging polymer for AM uses is PVC, with the clogging in the nozzle phenomenon observed. Diisononyl phthalate (DINP) plasticizer in concentrations up to 40% used in PVC compounds helps counteract the clogging phenomenon in a temperature range of 190 °C–210 °C, allowing for the production of AM structures with high flexibility [95].

Table 5. Research assessing printability of PEs and PEs composites.

Matrix Polymer	Fillers	Weight Fractions	Printing Method	Main Findings	Reference
rHDPE	Waste CFs	11.2–19.5 wt.%	FDM	Adequate crystallinity with an increase in mechanical response	[42]
	MWCNTs	0.5–5 wt.%	FFF	Higher melting temperature and better crystallinity	[46,90]
	Sawdust	45%	FDM	Dimensional deviation of 1–1.5% at 120 °C bed temperature and 188–198 °C nozzle temperature	[92]
rHDPE-rPP blend (70–30)	Biochar	20 wt.%	FFF	Biochar increased printability	[44]
rLDPE	SiC/Al_2O_3	40–50 wt.%	FDM	Improved wear properties	[48,49]
	MWCNTs	0.1–5 wt.%	FFF	Increased viscosity leads to better dimensional stability, mechanical recycling didn't affect the flowability of the composite	[94]

Polypropylene has a wide variety of applications, from high-volume manufacturing to applications demanding top-tier mechanical properties in the automotive industry. It has already been observed that blending recycled and virgin PP produces a rheologically suitable feedstock for 3D printing [55] but only rPP after thermomechanical recycling showed a drop in rheological and dimensional properties due to polymer chain scission due to thermal and mechanical stresses, oxidation, contamination and changes in the composition of additives [56,96]. The most common nozzle temperatures for rPP AM are around 200–250 °C [97], and for the first six recycling cycles, the onset degradation of the material starts at around 205–210 °C, crystallization temperature is at 116 °C and the peak degree of crystallinity is at 42%. Optimizing the printing procedure of rPP is vital for sustainable AM, since it has shown steady or slightly increasing mechanical properties from virgin PP over recycling cycles [58]. Polymer blends or recyclable thermoplastics (rPP-PET-PS blend), polymers that can be described as thermosets and thermoplastics (TPOs), or carbon (rCFs, CB)-reinforced PP showed a positive effect on improving the printability by lowering the degree of crystallinity in comparison to the rPP and elevating the glass transition temperature [54,98–100]. Natural fibers are already known for having a positive effect on the development of printable feedstock, and basalt fibers reinforced composite suitable for 3D printing have already been synthesized with the optimal weight fraction being 5% wt. [101,102], while cocoa beans shells used as filler helped reduce warping issues of rPP filament by 67% [103]. Tamarind fruit shells (TFS) and corn husk fibers (CHF) have been inserted into PP blends, but minimal reduction of warping was observed, and the maximum weight fraction reached was relatively low (3% for TFS, 7.5% for CHF). Reduced MFI and breakage issues due to air gaps were observed in higher concentrations [57,104]. Hemp and harakeke fibers composites have also been manufactured with harakeke-reinforced rPP (30% wt. harakeke fibers) having 84% less shrinkage than the original recycled feedstock [105]. An overview of the conducted research is stated in Table 6.

Table 6. Research assessing printability of rPP and rPP composites.

Matrix Polymer	Fillers	Weight Fractions	Printing Method	Main Findings	Reference
rPP	-	-	FFF	Stable morphological behavior and degree of crystallinity compared to virgin PP	[56,58]
	TFS	1.5–4.5 wt.%	FDM	Thermal stability at 230 °C adequate degree of crystallinity for 3D printing	[58]
	Bassalt	2–8 wt.%		High porosity compared to other conventional techniques for the same material, lowering of melting and degradation temperature in comparison to sole rPP	[101,102]
	CBS	5 wt.%	FFF	Reduced shrinkage and warping effects, decreased crystallinity, degradation were detected at 230 °C but did not affect the printing because of the small residue time	[103]
	CHF	2.5–7.5 wt.%		Elevating CHF concentration led to air gaps and voids. Printable up to 230 °C, reduced warpage, increased stiffness and rigidness	[104]
	Hemp, Harakeke	10–30 wt.%		Harakeke composite had shrinkage of 0.34%, rPP had 84%	[105]
	CB	0.5–10 wt.%		Reduced crystallinity on adequate levels for 3D printing, reduced warpage, MFI 13–19 g/10 min	[98]

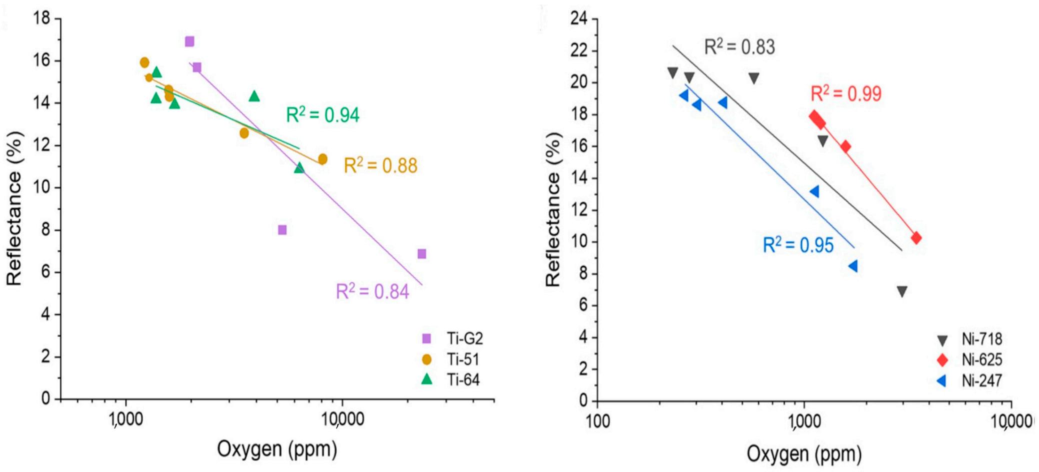
Other recycled polymers acquired from automotive waste are rABS and rPC, with the first one having proved to be suitable for AM processes. It was previously mentioned that rABS held a steady thermal and mechanical behavior over six recycling cycles [64] with thermogravimetric analysis (TGA) analysis showing that significant weight loss occurs at around 350 °C [61]. Multi-material structures with rABS, composites with metal powders or natural fiber composites have already been studied to improve mechanical performance depending on the wanted application without a negative impact on the thermal stability and printability of rABS [63,106,107]. Similar thermal behavior has been observed in recycled polystyrene (rPS) and polylactic acid (PLA) [108]. Using rABS as a filler (30% wt.) in rPC matrices helped improve dimensional stability and accuracy in the printing process, allowing for the exploitation of previously challenging materials for AM [109]. PLA is a natural polymer compatible with MEX printing either in filament form or in granule pellet form. Its suitability allows for the maximum valorization of its properties, providing a good candidate for AM processes [110]. The introduction of PLA-based composites aims at the replacement of conventional hard-to-utilize polymers with a more environmentally sustainable solution. Recycled carbon residues and recycled metallic powders seem to have a positive effect on improving specific static mechanical properties [66,68,69]. Thermoplastic polyurethane (TPU) and polyamide (PA) are materials that have been extensively researched in injection molding (IM) manufacturing but have not been utilized enough in AM. They have good flexibility and damping properties and are suitable materials for applications that require high energy absorption for safety issues. Elastic polyurethane foams have been produced by selective laser sintering (SLS) AM with the addition of TPU powder as filler enhancing the stability of the overall structure [65]. Fillers of GTR on PA and TPU bases have been studied with weight fractions up to 30% wt. The addition of this elastomeric filler had minimal effect on the stability of the PA composite but significant improvement of the TPU base by reducing the weight loss at the original degradation temperature range [111]. The overall efforts on the development of printable feedstocks exploiting elastomeric materials (TPU, PLA), and materials (rABS, rPC) are presented in Table 7.

Table 7. Other recycled polymers used in AM as matrix materials.

Matrix Polymer	Fillers	Weight Fractions	Printing Method	Main Findings	Reference
rABS	virgin ABS	0–50% wt.%	FFF	80–20 blend of recycled and virgin ABS had the same response as the sole virgin ABS	[61]
	-	-		Adequate thermal and mechanical behavior till the 5th recycling cycle, significant chemical degradation after the 5th cycle	[64]
	Fe powder	10 wt.%	FDM	Filaments produced with low speed had higher heat carrying capacity and less porosity.	[107,109]
rPC	rABS	30 wt.%	FGF	Increased printability on rPC/rABS blend than rPC at speeds of 30–40 mm/s	[106]
PU Foam	rTPU	2.5–7.5 wt.%	SLS	Increased compression strength by 60%, tear resistance by 31%, E modulus by 84% and better sound absorption	[65]
TPU	GTR	10–40 wt.%	SLS	GTR insertion decreased thermal stability, optimal weight fraction of GTR on 30%	[109]

3.1.2. Metal Additive Manufacturing

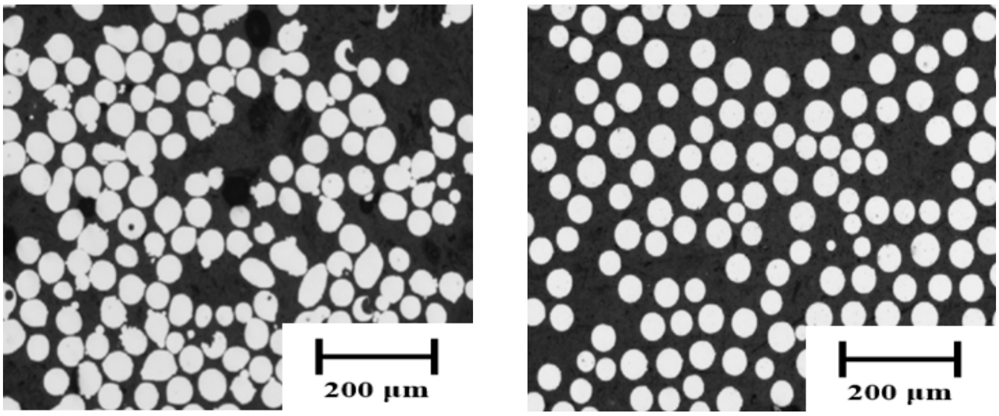
Due to their strength, metals are mainly used as fillers or sole materials and not in composites. Powder reuse is the main way of embodying AM in the recycling process. Metals are highly durable and strong, and metal powder can be reused for a significant number of cycles and is mainly used for PBF AM [74]. The main issues that have been observed are powder oxidation, which increases after every reuse cycle and hurts the formation of the printed specimens, and porosity increases leading to a sintering phenomenon between the powder's particles, affecting density and flowability [73,79,112]. An efficient way of assessing the changes to metal powder quality due to oxidation due to sintering or porosity changes is diffuse light stereoscopy (DLS). Gruber et al. [113] tested six different titanium- and nickel-based metal powders to investigate the connection between the percentage of the reflectance with the concentration of oxygen (Figure 4) via DLS and then with the degree of oxidation finding out that powders with a high degree of oxidation have low reflectance.



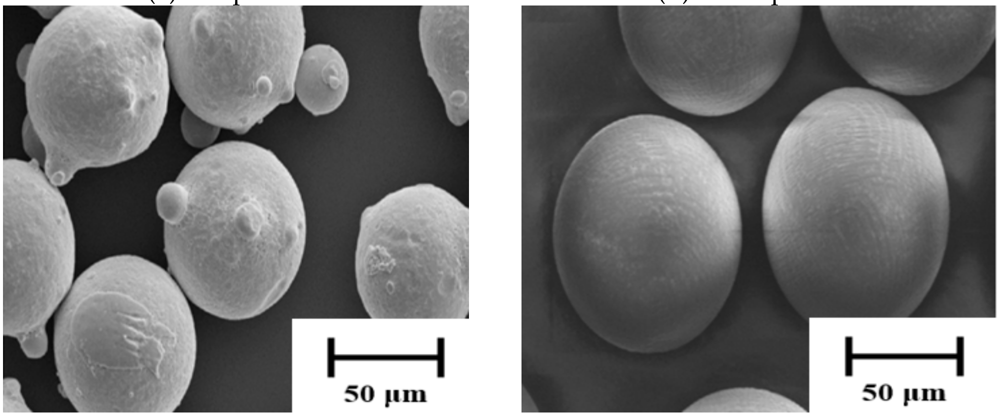
(a) (b)

Figure 4. Reflectance of oxidated degraded powders–oxygen concentration for (**a**) titanium-based alloys and (**b**) nickel-based alloys [113].

Aluminum alloy powders have not been extensively used in AM manufacturing because it is difficult to utilize aged or reused powders due to the significant effects of degradation on powder quality drop, leading to a big difference in comparison to virgin Al-based alloys. Although repeated AM processes are difficult with solely Al-based alloys, the acquired material could be used as a filler in other metals or composites. Bruzzo et al. [114] found out that heat treatment of aging aluminum alloys can lead to the production of a powder with equivalent quality and morphology to the original one, showing that future research could lead to promising results on the exploitation of Al-based alloys as a solution for sustainable laser metal deposition (LMD) AM. Recently, PREP has been introduced as an innovative way of producing improved quality metal powders. Chen et al. [115] made a comparison of nickel-based PREP-produced powders with GA powders, finding a recrystallization degree is higher in GA powders leading to lesser printing quality. Also, studies have shown that PREP as a process for recycling powders led to particles having a smoother surface, better microstructure and a reduced percentage of broken particles in the exported powder in comparison to the conventional GA process [77,78] as shown clearly in Figure 5.



(**a**) GA powder (**b**) PREP powder



(**c**) GA powder (**d**) PREP powder

Figure 5. Comparison of GA with PREP powder quality. (**a**) Metallographic cross-section of GA powder. (**b**) Metallographic cross-section of PREP powder. (**c**) SEM image of GA powder particles. (**d**) SEM image of PREP powder particles [77].

3.1.3. Additive Manufacturing of Ceramics

Clay is a ceramic that has great formidability and is easy to use in AM. It is also easily recyclable, since low energy is required for the process, and it is reusable, making it a good candidate for sustainable structures, as it has been shown that non-fired clay can be used for infinite recycling cycles, although there are some strict conditions regarding the contact of it with wet materials. It was noted also that for each recycling cycle, fresh water needs to be added to the mix [116]. It is vital to ensure that the amount of water is minimized for AM purposes, since too-high volumes could induce shrinkage and cracking phenomena during the drying process, but too low volumes should be avoided as well, to sustain the flowability and proper rheological properties. Jauk et al. [117] made a ceramic mixture of standard clay with a mixture of clay, water and wood sawdust, utilizing a twin-screw extruder for multi-material co-extrusion liquid deposition modeling (LDM) AM, making composites with gradient porosity and controllable humidity. Concrete is a high-strength ceramic material suitable for durable applications and efforts have been made to utilize it via material extrusion AM (FDM). Similar to clay, water-to-cement ratio plays a vital role in cracking phenomena. Also, the binder/aggregate ratio has been studied to be around 1:2–2.5 [80] and water to water-to-cement ratio around 1:0.3 [118] for achieving a printable cementitious feedstock. Flow direction has an instant effect on buildability by impacting the distribution and deformation of the supposed structure and interlayer bond strength. Pan et al. showed that these two cannot be improved simultaneously and stated the importance of flow direction optimization [119]. A ceramic mixture of cement glass with PET-G insertion as an aggregate has been studied as a possible alternative to the conventional bricks from the non-recyclable Portland-type cement and although a reduction in mechanical strength was observed, the composite had reduced specific heat and better thermal diffusion. Also, digital image correlation (DIC) studies have shown that the insertion of PET-G scaffold led to less abrupt failure. The final structure consisted of 85% recycled materials (75% ceramic, 10% PET-G) and had improved thermal properties regarding the printability of it in terms of decrease of thermal conductivity and increase in thermal diffusion [80,82]. Higher concentrations of PET (30–50%) led to further buildability increase, but problems have occurred due to possible detachments and the significant drop in strength stating that the optimal weight fraction is around 10% wt. [82]. Adding nano-biomaterials such as cellulose nanocrystals in small concentrations (1% wt.) helped access the printing problems of original reused cement by reducing porosity and densifying the microstructure leading to a better hydration degree. Also, inserting alkali-activating materials into Portland-type cement led to a significant drop in the environmental impact [120]. Besides concrete-based composites, earth-based structural material with significant concrete weight fractions and natural fiber traces limit shrinkage phenomena and improve the printability of the final composite [85].

3.2. Static Analysis, Impact Analysis and Dynamic Mechanical Analysis

3.2.1. Static Mechanical Analysis

The static behavior of materials is mainly examined from parameters such as yield strength, ultimate tensile strength (UTS) and Young's modulus (E), which can be accurately exported via tensile testing, allowing the identification of elastic, plastic region and fracture mechanisms. Plastics tend to have wide elastic regions, metals are more ductile, and ceramics are brittle materials. The form of the used feedstock (filament, pellet, granules) influences the tensile properties and the elastic, ductile or brittle behavior. For example, filament extrusion leads to materials with better surface finishing and dimensional accuracy resulting in better overall layer deposition and more consistent mechanical properties throughout the structure and a more ductile behavior. In contrast, pellets and granules do not present the same stable layer deposition, and this could result in flaws (air gaps) on the structure leading to deviations on the mechanical properties throughout the structure and a more brittle break mechanism. In general, filament extrusion is proposed for small-scale high-accuracy structures and pellet (or granular) extrusion for bulkier larger scale structures due to the fact that less time and cost are required for the process [35].

Another factor that determines the behavior of the structure is material selection. For example, rPET after the elastic region has almost brittle behavior in contrast to rPETG [121]. Bead orientation also influences the behavior of rPET, with vertical orientation making the extruded material more brittle [36]. Furthermore, thermomechanical processing leads to stiffening and strengthening with immediate effect on properties [38]. Printing quality parameters such as crystallinity influence porosity which could lead to breakage and demoted mechanical response [45,87]. Chain extenders (PMDA) and thermal modifier ethylene–ethyl–acrylate (EEA) slightly improved the mechanical response by around 15% without influencing stability, resulting in similar UTS to the traditional virgin PET (vPET stiffness around 45 MPa) [37,38]. Recycled carbon fibers have been used for producing composites with high strength; their effect on rPET showed a significant 390% improvement in tensile properties, greater than the conventional material at its first cycle of use [38]. The effect of the aforementioned fillers on tensile behavior compared to rPET is illustrated by the change of ultimate tensile strength (UTS) in Figure 6. Recycled carbon fibers had an equivalent effect in the rHDPE matrix at weight fractions up to 30% [40]. Carbon nanotubes (MWCNTs) showed even higher improvement in tensile strength (up to 100%) with significantly less weight fraction (5% wt.) [46,90]. MWCNTs filled in rLDPE helped strengthen the composite and also made it recyclable and manufacturable, but the constructed composite had lower values of UTS compared to the virgin counterpart (vPET around 25 MPa) [42,94]. Ceramic additions showed minimal effect and could not be added to high-weight fractions, with TiO_2 oxide leading to air gaps and breakage and hurting tensile testing compared to the original one [41], while SiC/Al_2O_3 filler showed a slight improvement in UTS end elongation at break [122], but further research about ceramic fillers needs to be conducted.

Significant research has been conducted on the effect of those fillers on the tensile behavior of rHDPE matrices compared to neat recycled ones and is illustrated by the change of ultimate tensile strength (UTS) in Figure 7.

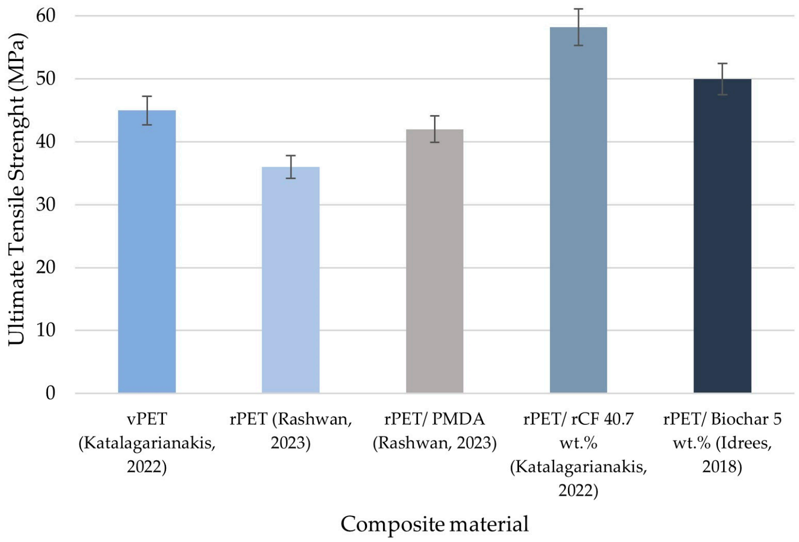


Figure 6. Fillers' effect on UTS for rPET matrices compared to the virgin counterpart [37,38,89].

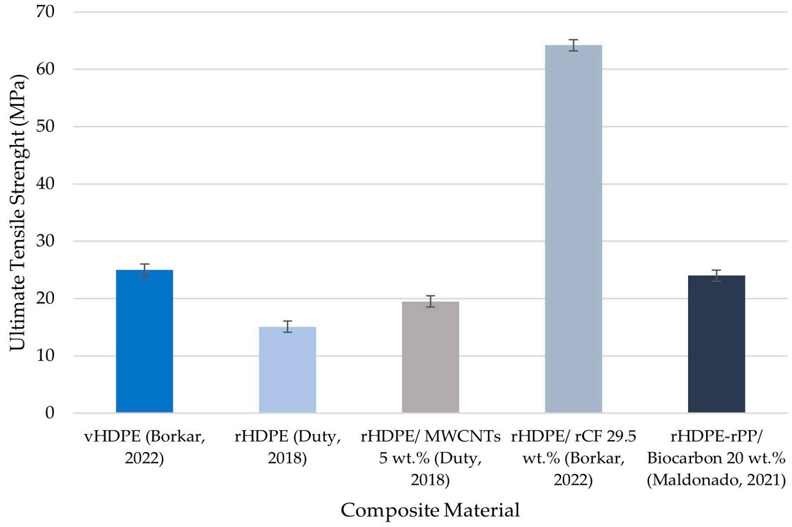


Figure 7. Effect of fillers on UTS of rHDPE matrices compared to the virgin counterpart [42,44,90].

Urban waste polymers are mainly easily recyclable, and a high volume of them is collected, but the applications after recycling/downgrading are limited. The need has emerged to recycle materials acquired from high-strength durable applications such as materials from automotive, aerospace, or construction applications (rPP), while tensile testing has shown a steady behavior on mechanical properties and acceptable changes in strength compared to virgin ones [55,56,58]. In comparison to conventional techniques, there is a significant drop in strength (20–40%), but the AM specimens show high ductility and good elongation at break contrary to the conventional IM technique [54,97]. Inserting fillers in rPP has helped solve printability issues and achieve tailored mechanical properties. Basalt fibers in a direction horizontal to the tensile test helped increase strength and close the difference gap to 15–28% in comparison to IM PP specimens. The direction of the inserted fibers after extrusion had an immediate effect on performance, with horizontal direction giving greater strength but angular direction increasing ductility and flexibility [101,102]. Natural fibers are an excellent alternative and a sustainable way to utilize agricultural waste. Tamarin fruit shells (TFS), corn husk fibers (CHS) and cocoa bean shells (CBS) were inserted in low-weight fractions and had minimal or negative effects in comparison to pure rPP on mechanical properties [57,103,104]. It is important to achieve higher weight fractions for better mechanical performance. Examples of natural fillers that had a significant effect on the tensile test were hemp and harakeke fibers with weight fractions around 30% and strength improvement at 70–75% [105]. The percentile effect on UTS of the inserted fillers is presented in Figure 8. On Figure 8a, the fillers that had positive effect on the mechanical properties comparing to conventional PP are presented and on the second section Figure 8b the reinforcements that caused drop on the mechanical properties in comparison to virgin PP are presented.

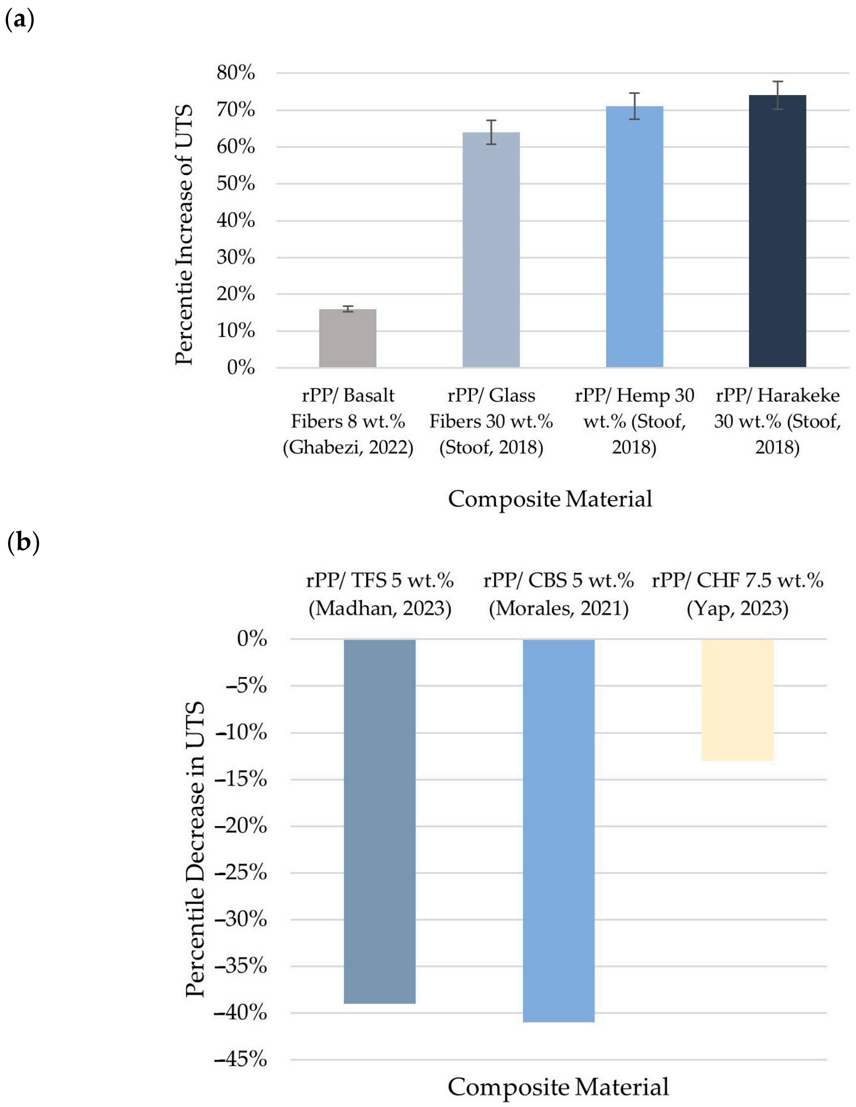


Figure 8. Filler's percentile effect on UTS of rPP composites. (**a**) Increase of UTS [101,105]. (**b**) Decrease of UTS [57,103,104].

Reused ABS had a decrease of 10–15% in comparison to virgin one which classifies as adequate, improved flexibility [59,60] and was studied to have a stable behavior 5 recycling cycles [64]. Polymers that require high levels of energy for the degradation process are stated as difficult to recycle, as PC or PS are primary examples of the aforementioned plastics and are mainly utilized in polymer blends with adequate properties, since the exploitation of them in additive manufacturing as sole materials was challenging due to the high temperature demands occurred from the difficulty of melting and degrading. It was found that the rABS/PC blend showed no decomposition or quality drop in weather exposure, making it a sustainable solution for outdoor applications [102], while recycled ABS/PS/PLA allowed for property optimization and had great load-bearing stability [108,123]. Metal powder-reinforcement rABS had around 100% more UTS than the plain matrix polymer [109]. The optimization of printing parameters and direction also has been stated, since the parallel distribution of the filler seems to have the biggest impact on strengthening the final composite [107]. Ground tire rubber-acquired materials seem to be great filler candidates for enhanced damping and energy absorption (260% increase), adequate for applications in safety equipment and automotive applications [124]. PLA and TPU are two interesting cases of thermoplastic with high elasticity, although their strength is lesser compared to petroleum-based polymers. The addition of biochar in relatively low weigh fractions (0.5% wt.) resulted in a 5% increase [66] in tensile properties, while rCFs increased strength by 500% in high-weight fraction CFs with relatively good recovery of the used raw materials (100% for rCFs and 73% for PLA), proving that recyclability and exploitation of CFs are possible and could be further researched [68]. Nickel-based reinforcements helped improve printing quality, but high-weight fractions caused significant negative effects on mechanical properties due to plasticizing phenomena [69]. TPU inserted in foams helped drastically improve foams compression strength while keeping an adequate elasticity behavior. Combining TPU with GTR waste showed an increase in porosity, causing a significant drop in mechanical strength, but alternative thermoplastic elastomers such as PA had more suitable properties [111].

For metallic materials, it has been observed that a steady static behavior exists regardless of the number of reuses, especially in titanium [74], steel [75] and nickel-based powders [76,115]. In aluminum alloys, there has been observed a moderate drop in mechanical properties with powder reuse [114]. Tensile tests for different powder production processes showed that PREP powders have lesser mechanical performance than GA powders, but they are better for AM processes due to their structural and thermal stability with immediate effect on printing accuracy [76,115]. The effect of powder reuse of steel alloys and aluminum alloys is presented in Figure 9, showing the difference in mechanical response of aluminum alloys compared to other metal alloys by detecting a drop of 31% on UTS for aged or reused powders. Another factor that determines the printing process in SLM AM; the dimensional accuracy and stability and directly influences material properties and the structures mechanical response is the volumetric energy density (VED). Pechlivani et al. [125] assessed the influence of VED during SLM processing on steel and cobalt–chromium alloys and found out that increase of VED lead to increased stiffness.

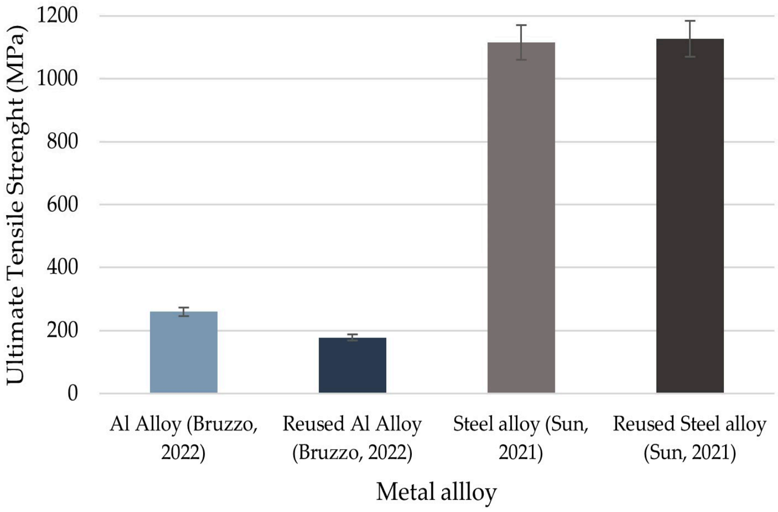


Figure 9. Effect of powder reuse on UTS on steel and aluminum alloys [75,114].

Concrete-based composites have been tested, with natural fibers or dust having a significant negative effect on mechanical properties after static tests [85]. The insertion of rPET scaffolding in concrete was shown to improve or maintain compressive static properties but direction had an immediate effect on mechanical strength with changing from horizontal to vertical resulting in a 50% decrease of the compressive strength. Also, high-weight fractions of rPET had an important impact on performance drop. The range of rPET reinforcement can help construct materials with tailored mechanical behavior [80–82]. Another way to achieve tailored mechanical properties has been proved to be topological optimization with Zhao et al. [84] studying the effect of different lattice structures on concrete-based composites and managing to construct composites with negative Poisson ratio leading to high energy absorption. Biomaterials such as cellulose nanocrystals seemed to be the optimal reinforcement for high-performance concrete-based materials with weight fractions at around 1% wt. resulting in a 15% static compressive strength increase [120].

3.2.2. Impact Behavior and Dynamic Mechanical Analysis

Besides standard static testing of materials, impact testing is used to determine important properties of materials for real-life applications. The quantification of impact strength (is used to measure the energy absorption ability of the specific material in a more realistic simulation of the actual circumstances. Izod impact testing is used for analyzing the behavior of brittle materials, especially for ceramics while Charpy impact is mainly used for metals which by their nature tend to be more ductile. For plastic materials, both tests have been exploited. Drop impact testing is used for identifying the energy absorption rates of materials in shock conditions.

In the literature, little research has applied impact tests to AM-produced recycled materials. Rashwan et al. conducted an Izod impact test on rPET matrices with PMDA and EEA and found an important elevation of the impact energy on the rPET/PMDA/EEA composite in comparison to plain rPET and rPET/PMDA matrix, making EEA a suitable candidate for impact modifying and showing the importance of research for appropriate reinforcements for the adequate utilization of rPET [37]. Charpy impact test of PETG showed a significant decrease in impact strength for each recycling cycle although static properties had a more stable response, meaning that stiffening after the continuous thermomechanical treatment led to a brittle plastic [39]. In contrary to PET or PETG, rPP and rABS presented a steadier behavior over multiple recycling cycles [58,64]. The effect of the printing angle on the impact strength was also analyzed, with vertical or horizontal orientation leading to more brittle behavior in comparison to angular (45°) [63]. Reused carbon fibers, although they had a significant effect on tensile strength, provided minimal improvement to impact properties (from 34 to 38 kJ/m^2) as it was studied in PLA matrices [68]. Biochar additives though helped improve impact strength by 140% with static properties improvement similar to recycled CFs making the valorization of biomass waste a promising field for further study [66]. Islam et al. [126] studied the insertion of rPP into concrete-based composite as an aggregate and examined the static and dynamic behavior of the structure. They found out that 5 wt%. insertion of PP aggregate resulted in a slight increase in compressive strength (11%) and a significant 68% impact energy drop at room temperature on the same weight fraction. In Table 8, a brief overview of the existing research regarding the mechanical response of recycled materials or composites is presented.

Table 8. Overview of the literature regarding the impact testing of recycled materials.

Impact Test	Matrix Material	Reinforcement	Weight Fraction	Main Findings	Reference
Charpy	rPETG	-	-	Brittle behavior of rPETG after the 3rd recycling cycle due to the continuous thermomechanical processing	[39]
	rPP			Steady impact strength over 6 recycling cycles	[58]
	rABS				[64]
	PLA	Biochar	4 wt.%	140% increase of impact strength on the composite	[66]

Table 8. *Cont.*

Impact Test	Matrix Material	Reinforcement	Weight Fraction	Main Findings	Reference
Izod	rABS	GTR	15 wt.%	Adequate impact strength resulting in high elongation at break (ductile behavior)	[63]
	rPET	PMDA	5 wt. %	Increase of impact strength from 36 MPa to 43 Mpa	[37]
Drop	Reused Concrete	rPP aggregate	5 wt.%	Decrease of impact strength by 68%	[126]

Dynamic Mechanical Analysis (DMA) allows the quantification of mechanical properties on dynamic phenomena as functions of time, temperature or frequency and helps provides a better overview of the material response on more realistic conditions than the conventional static testing. Recently there has been limited research and application of DMA on AM of recycled materials but it is adequate for viscoelastic materials and elastomers or composites making it a suitable analysis for a deeper understanding of the behavior of recycled polymeric composites. The calculations of storage modulus and loss modulus can help identify the damping behavior of the structure related to vibration and energy absorption. Frequency-dependent DMA for rPET and PETG showed a peak in loss modulus and damping behavior determining the glass transition temperature (T_g). Storage modulus had a dropping behavior with the rising of the temperature. It was also noted that lowering the layer thickness influenced the response of the specimen in DMA because of the better interfacial bonding [127]. Biochar insertion at relatively low weight fraction (0.5 wt.%) helped increase the PET composites storage modulus by 18% and on 5 wt. % weight fraction 42% increase was observed compared to the conventional one. Regarding the damping properties of the structure, increasing of the biochar concentration led to decrease of the tanδ meaning less damping and less energy absorption [89]. Temperature-dependent DMA on rPET and rTPU stated the effect of recrystallization on storage modulus after the glass transition temperature [128]. Frequency-dependent isothermal DMA on the glass transition temperature territory showed an effect of time on storage modulus, leading to the importance of both time and intensity of a specific phenomenon on material properties [129]. In the rPP matrix, CB had a positive effect on increasing the storage modulus by 21% on 10%wt. of reinforcement [95]. TPOs at high weight fractions up to 40% wt. on rPP matrix showed similar DMA properties as plain PP allowing the introduction of a polycrystalline multi-material that could work as a thermoplastic and thermoset for a variety of applications [100]. Identifying crashworthiness of materials is vital for automotive applications. Modal analysis of a rABS matrix showed that the insertion of recycled tire rubber at high weight fractions (50 wt.%) could lead to 260% improvement of damping properties [124]. In Table 9, a brief overview of the existing research regarding the DMA and the modal analysis of recycled materials or composites is presented.

Table 9. Overview of the literature regarding the DMA and modal analysis of recycled materials.

Test	Matrix Material	Reinforcement	Weight Fraction	Main Findings	Reference
DMA	rPETG	-	-	Temperature increase leads to decrease of storage modulus, peak damping properties on T_g	[127]
	rPET			Increase in storage modulus with temperature increase till the peak at T_g	[129]
		Biochar	5 wt.%	42% increase in storage modulus on the stated weight fraction, decreased energy absorption (72% damping factor for neat PET and 30% for the sustainable composite)	[89]
	rPP	CB	10 wt.%	Increased storages modulus (21%) and damping factor (4.45% for neat PP, 11.89% for the sustainable composite)	[98]

Table 9. *Cont.*

Test	Matrix Material	Reinforcement	Weight Fraction	Main Findings	Reference
	rTPU	PU foam	10 wt.%	Decreased T_g point (from 3 °C to −5 °C). Decreased damping properties (around 45% damping factor for neat TPU, less than 40% on the composite)	[128]
Modal analysis	rABS	GTR	50 wt.%	260% increase in damping properties after vibration testing (0.77% for neat ABS, 1.07% for ABS-10 wt.% GTR, 2.78% for ABS-50 wt.% GTR)	[124]

3.3. Life Cycle Assessment Analysis

Decreasing carbon footprint is vital for sustainable manufacturing. Additive manufacturing has proved to be more efficient than plain methods like injection molding for complex structures and batch materials with low amounts of different parts by having less impact on the global warming potential (GWP) and cumulative energy demand (CED) during the manufacturing process. The largest impacts are observed in the printing stage and the plate heating stage [130]. Life cycle assessment (LCA) analysis has shown that utilizing recycling materials for daily high-volume applications can lead to 80–90% less energy demand and 70–75% less carbon emissions compared to virgin PET or HDPE polymers [37,43]. Another alternative solution for efficient sustainable liquid packaging seems to be ENSO resins which were tested to be the optimal solution in terms of environmental impact [131]. Efficient valorization of e-waste plastics could be beneficial for acquiring recycled polymers with great elasticity (rABS). LCA analysis showed 28% fewer carbon emissions for the waste-to-manufacturing process compared to the virgin one, while the addition of rCFs also had a positive 22% reduction compared to the equivalent plain ABS [60,132,133]. In general, recycled ABS led to solid amounts of hazardous particles during the printing process, a phenomenon that has been compressed with the use of PLA for AM processes [134]. LCA analysis of PLA recycling stated that there is 97% less impact in the aforementioned categories but in France and USA increased impact on the Ion radiation was observed due to the energy generation from nuclear plants in these regions [135]. Further research on the effect of recycling on environmental impact must be conducted to address the embodiment of polymeric composites in AM. Biomass waste as a filler in ceramic matrices was shown to reduce CO_2 emissions by 43% [66]. LCA analysis in metal powders has shown that the stage with the biggest impact is the atomization stage, due to the electricity demand and the high consumption of argon noble gas. Embodying novel methods such as plasma atomization could lead to a more sustainable approach to metal AM [136]. The stage of feedstock production seems to be the most energy-demanding in ceramics as well. AM can emerge as an efficient way of utilizing cementitious composites. The insertion of natural fibers (rice husk) can lead to a significant decrease in environmental impact. Also, a 3D printing wind turbine made from concrete showed 16–24% less emissions of carbon dioxide in comparison to the original steel one but the energy demands were significantly higher for the process (29% for normal strength concrete, 64% for high strength) [85,137].

4. Large Scale Additive Manufacturing and Possible Future Applications

Currently, AM has been mainly used for small scale, low volume manufacturing and prototyping. Design freedom and reduction of material waste are significant advantages of AM in comparison to other conventional manufacturing processes. Embodying AM with larger scale manufacturing, by developing printable composites and multi-materials can lead to an efficient exploitation of reused materials leading to a circular economy model [43]. Adding the right fillers in polymeric matrices proved to help achieve tailored properties. Carbon and glass fibers inserted on recycled polymers helped greatly improve static properties and lower the coefficient of thermal expansion (CTE) to similar levels to concrete providing an alternative for structural applications [138]. The addition of polymeric (rPET) scaffolding on a ceramic base led to a reduction in mechanical properties but led to a brick composite that could be easily printable making it appropriate for low-demanding structural applications [80–82]. Finite element analysis (FEA) showed that the construction of a 3D printed roof made from rHDPE without steel reinforcements is feasible and a lab scale manufacturing prototype was constructed, allowing room for further research about the optimization of sustainability in the construction industry [139]. Besides structural applications, manufacturing-as-a-service (MaaS) can help expand the application range by providing better flexibility in design customization and scalability. Automotive and aerospace, medical or biomedical, and the production of everyday equipment (e.g., sports) could be possible examples of the introduction of MaaS in the industry [140]. Hybrid polymeric composites utilizing recycled carbon or glass fibers have already been shown to be a great solution for lightweight durable automotive components [141]. Carbon black reinforced polypropylene was clarified as an adequate alternative for repairing automotive headlights [98,142]. Agricultural products from AM have been constructed with prime examples being deployable modular farms out of rPET [143], water-oil separation devices out of ceramic matrices with in-situ grown whiskers [144] and for Internet of Things (IoT) agricultural toolboxes designed for better soil and environmental monitoring [145]. AM has shown great potential for utilizing recycling feedstock for sports equipment or clothing. TPU and PLA have been used lately to produce 3D printed shoe soles [128,146] and other flexible products (e.g., protective mouthguards) [147]. Protective sports equipment is an adequate application for reused automotive and electronic waste polymers (rABS or rPC). Romani et al. [106] managed to construct 3D printed shin pads out of the aforementioned recycled polymers. ABS and PLA have shown adequate electrodynamical properties making them suitable candidates for electrochemical sensors used in analytical chemistry [148–150]. 3D printing of catalysts for microwave-assisted reactions was being researched by Tubio et al. [151] by utilizing a palladium-alumina composite that could handle 200 cycles of reuse. AM can coexist with conventional techniques (Injection Molding). Recycled ABS has been utilized to produce molds that have been later used for investment casting [152]. Assisted investment casting could be an efficient way of exploiting rLDPE as well, a material with low viscosity and low melting point [50]. Finally, AM is adequate for applications in the biomedical industry. Appropriate topological optimization was used to achieve tailored mechanical properties on nickel-based alloy 718 for orthopedic applications. Finite element analysis showed that adequate implants can be constructed with Additive Manufacturing techniques [153,154], with a tibial implant prototype already been constructed by utilizing SLS AM [155].

5. Research Gaps and Solutions

Future studies could focus on incorporating additional materials into Additive manufacturing such as recycled Polycarbonate, Ethylene Vinil Acetate (EVA, ground tire rubber waste) and Nylon (urban and industrial waste). These materials are available in high volumes, yet there is a lack of research on the integration of them in additive manufacturing, or exploring new composites with recycled ABS, PLA and TPU matrices, materials that have been proved to be adequate for additive manufacturing for achieving tailored mechanical properties and expanding the range of possible applications. Biomass waste fillers (biochar, carbon black) could work as an alternative to conventional carbon fibers and further research needs to be conducted to establish them as a potential replacement. Concrete or Metal printing and the exploitation of construction wastes can also be a possible aspect of future research in terms of raw material acquisition not only as matrix materials but as possible fillers as well. Due to their applications, limited research has been done to metal recycling and because of the printability issues there is a lack of research for utilization of recycled ceramic materials. Natural fibers from agricultural waste and specific nanomaterials and the effect of those fillers on improving the printability and performance of otherwise difficult-to-utilize polymers (PP, PEs), can help embody these materials in AM processes as well and needs to be further searched, along with the identification of the optimal weight fractions. Besides new materials, optimization of the production process of printable feedstock can also be searched with novel processes (PREP atomization) emerging for processing metal powders. A lack of research regarding impact testing and dynamic mechanical analysis testing has also been observed, analyzing the dynamic behavior of an advanced material is vital for upscaling and applying it in real life applications. Drop impact testing can help identify properties like energy absorption on shock-dynamic conditions while DMA allows the identification of storage modulus and damping factor of materials, quantifying the vibrational behavior of a material, vital for applications that require identification of the crashworthiness in automotive, structural, or aerospace industry. Previous LCA analyses have shown that AM has the potential to reduce environmental impact and can be used as a step towards to circular economy, but further research needs to be conducted for more recycled materials. Assessing the environmental impact of the recycling process from waste to feedstock for more materials, especially for industrial polymers, reused Metals or Ceramics from structural applications, can help achieve a net zero waste approach, beneficial in the long term.

6. Conclusions

In the present study, a comprehensive review of the research on the utilization of end-of-life products through AM processes has been conducted. The integration of a circular economy model and the sustainability factor are vital for the viability of the manufacturing industry. AM has emerged as an efficient way of utilizing reused materials due to its flexibility and reduced waste. Especially for polymers, recycled feedstock is available on the market for automotive or e-waste plastics but due to the difficulty of the procedure, the steps were kept confidential. For recycled polyethylenes, the waste-to-feedstock process is relatively easy, since they have low amounts of energy demands for the recycling process making and specific uses on the first cycle of life; many studies focused on the recycling process of polyethylenes along with the mechanical characterization. Given that recycled materials tend to exhibit less strength than their virgin counterparts, there is an emerging need for advanced composite materials particularly for polymer-based composites. The insertion of composites into AM is primarily feasible through MEX 3D printing. However, recent efforts have explored the use of polymeric feedstock, particularly elastomers such as TPU, reinforced with fibers in powder form for SLS AM (Flexa powder-TPU with CFs). Determining the optimal weight fractions for possible fillers is another aspect that can lead to the production of efficient novel composites. Custom-made feedstock is easier to be inserted in MEX AM. Optimizing the printing process has an immediate effect on the mechanical response of the printed composite. Static behavior analysis provides a comprehensive view of the mechanical response for novel non-tested materials while dynamic test such as impact or dynamic mechanical analysis can be used for the transition for larger scale additive manufacturing of already tested materials. Materials with high damping properties can be efficiently utilized by the automotive industry due to their high energy and vibrations absorption. Also, it is important to take into consideration the environmental impact additive manufacturing has and compare it to other conventional manufacturing processes. Carbon emissions, energy demands and material usage are all factors that can be calculated via appropriate Life Cycle Assessment analysis which is vital to ensure that Additive Manufacturing can be used as sustainable technique that promotes circular economy. In conclusion, in order to upscale the AM processes for large-scale applications, more comprehensive research regarding the dynamic response of advanced materials and the assessment of the environmental impact needs to be conducted.

Author Contributions: Conceptualization, I.F.K., N.K. and K.T.; methodology, I.F.K. and N.K.; validation, I.F.K., N.K. and K.T.; investigation, I.F.K. and N.K.; writing—original draft preparation, I.F.K.; writing—review and editing, N.K., E.M.P. and K.T.; visualization, I.F.K.; supervision, N.K., E.M.P. and K.T.; project administration, E.M.P. and K.T. All authors have read and agreed to the published version of the manuscript.

Funding: This research received no external funding.

Institutional Review Board Statement: Not applicable.

Informed Consent Statement: Not applicable.

Data Availability Statement: No new data were created or analyzed in this study.

Conflicts of Interest: The authors declare no conflicts of interest.

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