Author Accepted Manuscript (AAM)
This is the link to the accepted manuscript version of the article published in *Journal of Cleaner Production*, Volume 386 (2023), Article 135779.

The final published version is available at: https://doi.org/10.1016/j.jclepro.2022.135779

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Biomass waste materials through extrusion-based additive manufacturing: a systematic literature review

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Abstract

Circular economy and bioeconomy models have increasingly spread the principles of sustainable development through different strategies such as reuse and recycling. Biomass waste and by-products are often considered valuable resources. Digital technologies, i.e., extrusion-based additive manufacturing, may potentially foster their exploitation as new materials, although the current framework has not been clearly defined yet. This work wants to systematically review the publications focused on new materials from biomass waste or by-products for extrusion-based additive manufacturing processes. The current situation was analyzed on 69 selected works from 2016 searched on Scopus and WoS, focusing on the different raw materials, new 3D printable materials, 3D printing processes, and potential applications. Afterward, the emerging trends were highlighted through selected best practices from the design practice and industrial sectors. Despite the significant development of thermoplastic reinforced materials, i.e., PLA-filled composites, a wide range of biomass waste and by-products have been studied, especially for small format low-cost Fused Filament Fabrication. Although the academic field may be less focused on exploiting these new circular materials, the interest in new applications is increasing within the design practice and industrial sectors, fostering new synergies within bioeconomy and circular economy contexts.

Keywords: Additive manufacturing; Design for sustainability; Circular economy; Upcycling; Fused Deposition Modeling; Direct Ink Writing;

1. Introduction

In the last decade, the new economic development model called circular economy (CE) has gained ever-increasing attention due to the need to minimize the environmental impacts of human activities and its potential to promote well-being and a harmonious society [1]. CE provides a novel concept of economic systems, enabled by several national governments worldwide, aiming to maintain extraction rates of resources and preserve their value as long as possible within the system [2]. The CE concept aims to accomplish sustainable development, as opposed to the traditional linear models of production and consumption [3,4]. Current linear systems follow an extract-produce-use-dump material and energy flow model and expose many companies to risks, such as rising resource prices and supply disruptions [5]. One of the principles on which CE is based is to keep products and materials in use through several strategies, i.e., by maximizing resource recovery with reusing and recycling [2,6,7]. Consequently, the value of waste is maintained as long as possible, viewing it as a resource to be reused and reintroduced into the economic system. Waste management plays a crucial role in the CE since it facilitates the transformation of biomass waste or by-products into raw materials [8]. However, there are still some limitations and challenges to be addressed to reach the potential of CE [9].

Different strategies may be intertwined to boost the transition toward the CE. In particular, bioeconomy can be beneficial because bioeconomy can be considered, to some extent, intrinsically circular, exploiting input materials and resources, which are bio-based and renewable [10,11]. Bioeconomy indeed aims at using renewable biological resources from biomass rather than fossil- and mineral-based resources to produce goods, foods, and energy [12]. However, new business models must be designed, implemented, and managed to make the bioeconomy more circular and expand its potential [10]. To this end, the valorization of biomass and bio-based waste through different strategies and technologies has been the aim of several research works that have been pursued in the last decade [13]. Among these, some approaches can be identified as very popular such as the incorporation of functionalized lignin in polymeric materials [14], biological and thermochemical treatments of bio-based waste coming from domestic, agricultural, and industrial activities [15], and the production of functional bioplastics and advanced materials from agri-food waste [16]. Moreover, some key enabling

technologies (KETs) have been identified as more important and promising than others, such as genetics and bio-refineries, because they have the potential to fulfill more criteria perceived as crucial for the transition toward a bioeconomy [17]. The transition to bioeconomy is, in fact, hindered by many factors, such as the low competitiveness of bio-based products and the extensive use of edible crops, causing detrimental effects on the environment without economic profitability. For these reasons, it is helpful to identify which KETs can address these limitations of bioeconomy and support the transition toward a more sustainable system.

Within this framework, additive manufacturing (AM) stands out as being one of the most used technologies to foster new circular economy models [18–20]. According to the international standard (ISO), AM is defined as a class of processes able to obtain objects from 3D model data through a layer-by-layer approach, different from subtractive manufacturing and formative manufacturing methods [21]. Additive technologies or 3D printing represent an essential innovation nowadays in different sectors, offering several advantages compared to conventional subtractive methods. The main important ones are the capability to realize more complex geometries, the possibility of printing various materials (e.g., polymers, metals, ceramics, etc.), the waste minimization, the ease of personalization of products for small-volume productions, and the possible implementation of a distributed network of low-cost reprocessing machinery in local communities [22]. For these reasons, it is considered the key to the fourth industrial revolution, based on intelligent automation technologies and smart factories [23]. Moreover, the role of AM in promoting a more sustainable economic system is emerging, considering its significant benefits and increasing applications [24]. For instance, AM has been recently used for the recycling of a wide range of resources at their End-of-Life (EoL), such as electronic waste [25], thermoplastic polymers [26], materials from the construction sector, and rubber tires [18]. In addition to waste minimization and reduction of material consumption, AM can also foster the use of waste biomass, avoiding the detrimental use of edible crops as done so far to obtain most input materials. Furthermore, AM can limit pollution due to landfilled agro-waste and waste materials produced during conventional manufacturing processes. All these aspects tackle some environmental issues, which hinder the development of the bioeconomy and which AM processes can address to accelerate the transition towards a circular bioeconomy. Also, social issues can be answered by AM, which has already contributed to job creation in Sweden, particularly in product development stages with rapid prototyping and production stages with the manufacturing of low-volume complex design products [27]. To this end, AM combined with waste biomass shows the potential to foster the use of eco-design strategies, i.e., designing and fabricating long-lasting products by upcycling agro-waste, enlarging the possible exploitation in real contexts [11,19]. Additive technologies mainly involved in a CE perspective are the "Material Extrusion" technologies or extrusion-based AM, thanks to their accessibility and flexibility [19,21]. Fused deposition modeling (FDM), also known as fused filament fabrication (FFF), and direct ink writing (DIW), also called liquid deposition modeling (LDM), represent the main extrusion-based AM processes. From the literature, new circular materials and applications have been developed, starting from FFF and DIW technologies. For instance, the former enabled the recycling of biodegradable and synthetic polymers [28–31], while the latter the recycling of fiber-reinforced polymers and ceramics [32,33]. However, how extrusion-based AM technologies have been employed so far to recycle bio-based waste is still unclear. To this end, a particular focus on bio-based waste materials, which is a subset of the entire collection of bio-based materials, and its application in AM context is crucial to support the transition to a more sustainable circular bioeconomy.

This work, therefore, focuses on the systematic review of the works related to new materials from biomass, either waste or by-products, for extrusion-based AM processes. Three main research questions (RQs) were framed:

- RQ1: "What is the current situation in the academic field considering extrusion-based AM, materials science, and open and closed loops of biomass as raw materials?"
- RQ2: "What are the most used biomass raw materials, new 3D printable materials, and extrusion-based AM processes in this research context?"
- RQ3: "What are the emerging applications and trends, as well as the limitations and perspectives within this research context?"

After explaining the methodology, a general analysis of 69 works from 2016 is depicted to frame the current situation (RQ1). These works are then analyzed according to the raw materials, the new materials, and the extrusion-based AM processes (RQ2). Some considerations about the limits and future perspectives about this context are then made starting from the real and potential applications of these emerging materials, including some best practices from the design practice and industrial sectors (RQ3). Although applications are still limited, biomass as raw material can generate new synergies and virtuous cycles within the bioeconomy and CE contexts, especially at the intersection of design practice. This work wants, therefore, to stimulate further research and application-driven works to exploit these materials in the real world. Furthermore, this review aims to give a different and comprehensive perspective on this topic, differing from the other reviews because of the holistic consideration of the whole raw material lifecycle, which means from biomass waste to the potential applications, as well as its pragmatic approach to the topic.

Nomenclature

ABS	Acrylonitrile Butadiene Styrene	PBAT	Polybutylene Adipate
AM	Additive Manufacturing	PBS	Polybutylene Succinate
CE	Circular Economy	PCL	Polycaprolactone
CoPE	Copolyester	PE	Polyethylene
DIW	Direct Ink Writing	PHA	Polyhydroxyalkanoate
EoL	End-of-Life	PHB	Polyhydroxybutyrate
HDPE	High Density Polyethylene	PLA	Polylactic Acid
FDM	Fused Deposition Modeling	PLLA	Poly L lactic Acid
FFF	Fused Filament Fabrication	PP	Polypropylene
KET	Key Enabling Technologies	PTT	Polytrimethylene Terephthalate
LCA	Life Cycle Assessment	TPU	Thermoplastic Polyurethane
LDM	Liquid Deposition Modeling	TRL	Technology Readiness Level

2. Materials and Methods

This paper analyzes the works that use biomass waste and by-products as a starting point for new 3D printable materials through a systematic literature review following the PRISMA systematic review statement. This reporting guideline aims to improve the transparency, completeness, and accuracy of systematic reviews and meta-analyses. It mainly consists of a checklist and a flow diagram that help in verifying the exhaustivity of the information and showing the workflow behind the search, identification, screening, and analysis processes [34,35].

The search strategy is resumed in Table 1. In short, this work includes accessible research articles with an experimental part related to extrusion-based AM processes (Material Extrusion category from ASTM) [21], excluding reviews, meta-analyses, or works with a theoretical perspective on 3D printing. After a preliminary screening, a timeframe from 2015 to 2022 was set since no works were found before 2016. Since this review investigates the intersection between circular economy and bioeconomy, discarded articles deal with renewable or bio-based pristine materials or biomass without any clear evidence of their waste origin, either not clearly stated or not presumable from the text. Finally, works without the main details related to the AM process, i.e., the type of 3D printer, the type of biomass, or the newly developed materials, were excluded from this review. The review was conducted on Scopus and Web of Science (WoS) academic repositories from April to mid-December 2022, as reported in Table 1. Scopus and WoS databases were selected to avoid the presence of inconsistencies regarding content verification and quality. Similar search strategies were followed, starting from a set of common keywords and defining the two specific query strings. For WoS database, the query string did not include the keywords “Circular economy” OR “Bioeconomy,” unlike for Scopus database, because adding these keywords dramatically reduced the number of articles found in Wos database with respect to the results without these keywords.

Table 1. Eligibility criteria, search databases, and corresponding query strings for the literature review presented in this work.

Eligibility criteria	Search databases	Query string
• Accessible research articles. • Works with experimental activity on AM processes. • Works with experimental activity on biomass waste, scraps, and by-products. • Works focused on Material Extrusion AM (or extrusion-based AM) [21]. • Data completeness about AM processes, waste/by-products, and recycled materials	Scopus	("Additive manufactur*" OR "3d print*" OR "rapid prototyp*") AND ("agricult*" OR "agroindustr*" OR "agrifood" OR "agri-food" OR "agrofood" OR "agro-food") AND ("waste*" OR "scrap*" OR "biomass*") AND ("Circular Economy" OR “Bioeconomy”)
	Web of Science (WoS)	(((ALL=(Additive manufacturing OR 3D printing OR Rapid prototyping)) AND ALL=(agricultural OR agroindustrial OR agrifood OR agri-food OR agrofood OR agro-food)) AND ALL=(waste OR scrap OR biomass))

Search results were initially identified and screened according to their language (English), avoiding duplicates, retired or not accessible works. The screening started with a single review of titles, abstracts, and keywords, selecting the excluded and included records. Furthermore, publications that cited or were cited by the screened records were checked to include potential records that could meet the eligibility criteria but were excluded from the query strings. These results were then inserted into an excel matrix to preliminary screen the full texts according to the biomass, the raw material, and AM process. Two additional screenings were undertaken by carefully reading the full text and assessing the data completeness.

Data extraction was performed by filling the previous matrix and adding new data columns related to biomass, new material, AM process, and potential applications. Data were collected from the full text, the link of the publication, and,

in case of discrepancies, from other sources or websites, i.e., filament producers or waste suppliers. In some cases, data were also normalized to make them comparable, i.e., weight fractions. Afterward, data were analyzed using excel, creating a matrix to examine specific aspects, i.e., type of waste. This matrix was also used to plot the graphs to visualize the analysis results, and it is visible at: https://github.com/piuLAB-official/Dataset_A.Romani_2023_JCP. Some best cases from the design practice and industrial sectors were finally selected to deepen the discussion of the results from the literature review, trying to understand possible synergies between academic research and practical context. These case studies were searched in design project repositories or starting from the commercial solutions used in the experimental activities of the screened studies.

3. Results and Discussion

This section presents the analysis of the 69 studies from the systematic literature review process described before. Sub-section 3.1 relates to the general analysis and gives some insights on RQ1. Sub-sections 3.2, 3.3, and 3.4 are respectively focused on analyzing the raw materials, new materials, and the specifications of the 3D printing processes. Finally, sub-section 3.5 resumes the discussion on the emerging applications, answering RQ3.

Many works cited 3D printing technologies as possible future development without experimentation or only mentioned the possibility of using biomass waste and by-products as raw materials. Furthermore, some results focused on different research areas linked to AM, bioeconomy, and circular economy, i.e., Industry 4.0-related topics.

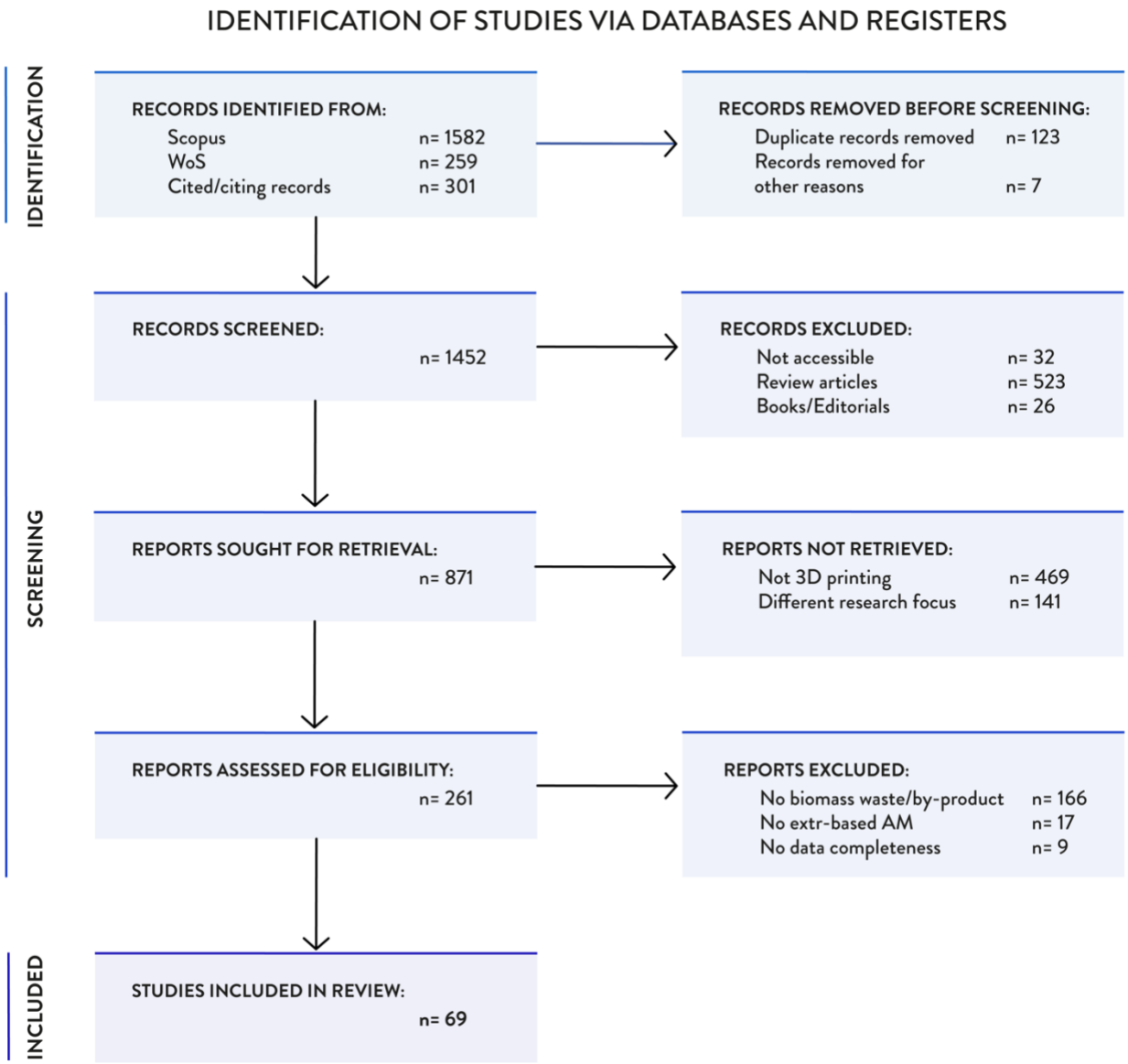


Fig. 1. PRISMA flow diagram showing the selection process of the literature review presented in this work.

3.1. General analysis

The general analysis aimed to frame the state of the art related to new 3D printable materials from biomass waste and by-products as main raw materials. As previously seen in Fig. 1, 69 works meet the eligibility criteria. Even if the timeframe of this review comprehends the last seven years, no significant publications were found before 2016. This fact could probably be linked to the real exploitation of extrusion-based AM processes, which only started after the expiry of the first patents [4]. According to Table 2 and Fig. 2a, the interest in developing new materials for 3D printing from biomass has been increasing since 2016, especially in the last four years. As a matter of fact, 13 works were counted between 2016 and 2018, whereas 56 works, about 80% of the results, were found from 2019 to 2022. Furthermore, the publications of 2022 may increase, considering the provisional number retrieved by this review. New works could be published in the last weeks of this year, and other publications may not be indexed within Scopus and WoS yet.

In general, only one conference paper has been found, showing that publications in academic journals are the most common way to disseminate these results. The publications were then analyzed considering their main research area, which means the primary and most representative one. In detail, the following aspects were considered to assess this aspect: the research field of the journal, the keywords of the specific works, and the contents of the research. The complete list of the journals is visible at: https://github.com/piuLAB-official/Dataset_A.Romani_2023_JCP.

Eleven clusters were detected from the analysis of the main research areas. In particular, one-third of them is directly related to the field of materials science. Three of them can be considered sub-fields, and they were defined considering the relatively high number of selected records ("materials science and engineering", "polymers science and engineering", "composite engineering", and "materials science for specific sectors"). The works of materials science-related research areas represent more than 55% of the publications, which means 39 works (Fig. 2b). Other research areas can be associated with manufacturing ("manufacturing and processing engineering", and "additive manufacturing"), sustainability ("environmental sciences", "waste management"), or general science ("chemistry and physics" and "applied sciences"), representing ~13%, ~13%, and ~10% of the works, respectively. Less represented sectors are related to bioengineering and bioprinting, which could also be linked to some works within the materials science-related clusters. Materials science appears as the most involved research area at the intersection of the topics of this review. However, there is an increasing interest from other disciplines, fostering collaborations and multidisciplinary projects, i.e., experimental activities for applications and demonstrations of different application fields such as architecture and product design ("materials science for specific sectors").

Table 2. List and number of the selected works according to the main research area and publication year.

Main research area	2016	2017	2018	2019	2020	2021	2022
Materials science and engineering	//	[36]	[37]	[38], [39], [40], [41], [42]	//	[43], [44]	[45]
Polymers science and engineering	//	//	[46]	[47], [48], [49]	[50], [51]	[52], [53], [54]	[55]
Composite engineering	//	[56], [57]	[58]	[59]	//	[60], [61], [62], [63], [64]	[65], [66], [67]
Materials science for specific sectors	[68]	//	[69]	[70], [71]	[72], [73]	//	[74]
Manufacturing and processing engineering	//	//	[75]	//	[76], [77], [78]	[79]	//
Environmental Sciences	//	//	//	[80], [81]	[82], [83]	[84]	//
Additive manufacturing	//	//	[85]	[86]	//	[87], [88]	//
Chemistry & Physics	//	//	//	[89], [90]	[91]	[92]	//
Applied sciences	//	//	[93]	//	[94], [95]	//	//
Waste management	//	//	//	[96]	//	[97], [98]	[99], [100]
Other	//	[101]	[102]	[103]	//	//	[104]

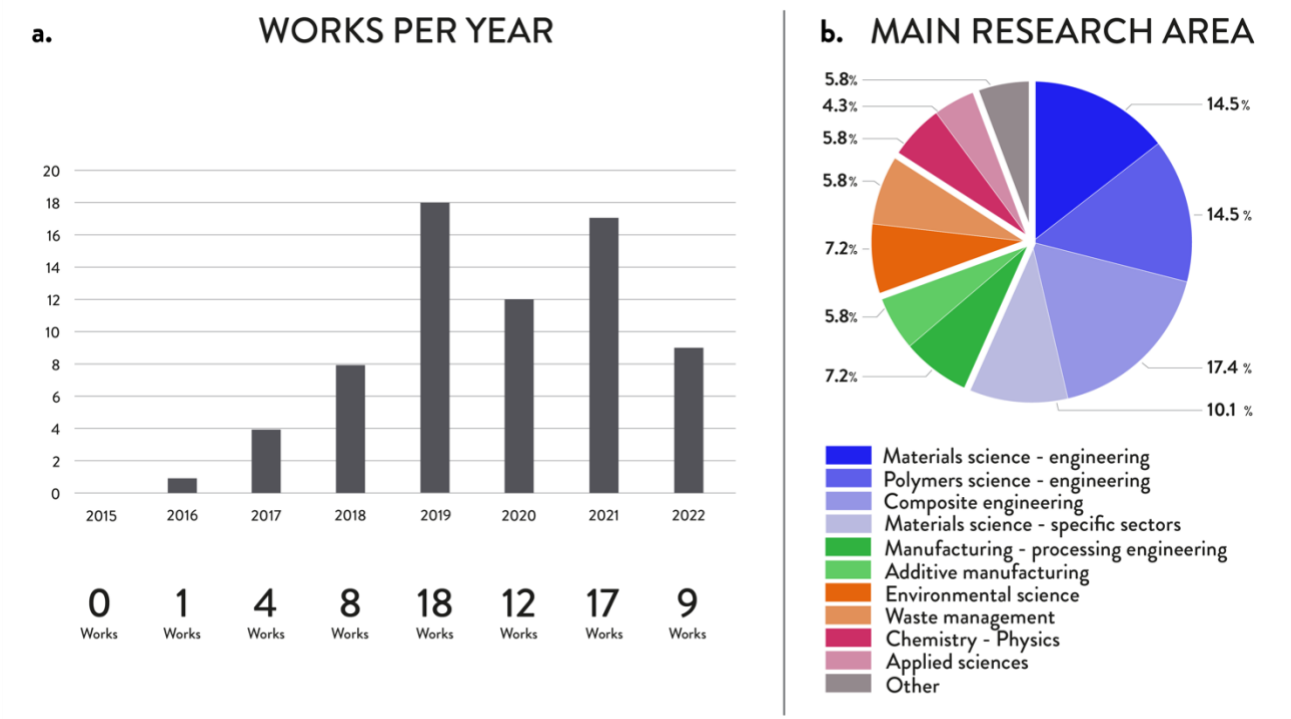


Fig. 2. General analysis: (a) works per year; (b) main research areas of the selected works.

The transition towards sustainable production and consumption often implies not only the environmental aspect but also different points of view, i.e., social ones [68–70]. The publications were reviewed to check whether they considered the environmental and social aspects through specific methodologies or procedures. Accordingly, only one publication included a Life Cycle Assessment (LCA). In this case, the work of Sinka et al. gives a first idea of the environmental impacts of the new material applied to the building sector [82]. The lack of LCA for 3D printable biomass materials may be due to the preliminary nature of the studies or a focus on the characterization of these new materials since LCA procedures generally consider practical applications and real case studies to set the correct functional units. Similarly, no social LCAs were found, and probably this issue is linked to the limited number of practical applications within these studies, as better explained in Sub-section 3.5.

3.2. Waste, scraps, by-products

This sub-section analyzes the bio-based waste used as raw materials within the selected works. As shown in Fig. 3a, biomass waste and by-products were obtained from industrial activities in most cases. The waste originated from end-users and consumers for nearly 14% of works. According to Fig. 3b, almost one-third of the works exploited wood and timber as waste material, mainly supplied as feedstock in the form of fibers and sawdust (Fig. 3c). Still considering the wood waste type, either wood flour [51,57,95] or wood chips [39,40] were provided as feedstocks only in a few cases (Table 3). In more than one-third of the works, no explicit indications were found for the wood classification. When specified, the pine tree is the most recycled wood type, followed by some examples of recycling for eucalyptus, spruce, and maple (Fig. 3d). Apart from wood waste, most of the selected works (~40%) have employed waste derived from the processing of herbaceous plants (Fig. 3b). Within the waste category of herbaceous plants, hemp was the most used as recycled feedstock in various forms for example shives [59,82,102]. Fig. 3e also shows that sugarcane in the form of bagasse [86] and rice husks [52,98] were also employed as a recycled herbaceous material. The rest of the works recycled waste and by-products of fruits and seeds (~19%) and paper pulp, fish, and mussels (~13%).

The shape and size of discontinuous fibers in wood-based composite materials are critical parameters for extrusion-based 3D printing processes because they strongly influence the physical and mechanical properties of the final polymer composites, especially those reinforced with natural fibers [105]. To improve the reinforcement dispersion in the extruded materials, natural fibers were usually ground and sieved, obtaining particles with dimensions suitable for 3D printing. For instance, before mixing with other 3D printable materials, a wood flour with a particle size lower than 125 µm was employed for an FFF printer [89] or even lower than 100 µm for DIW processes. A median size value of 75 µm was measured by Pitt et al. for wood flour used for DIW extrusion printing [57], while dimensions lower than 75 µm were also presented by Kam et al. for DIW technology [38]. In other cases, thermomechanical pulp fibers were milled to

achieve an average fiber length of 0.4 mm and a diameter of 38 μm [46]. Even though larger fiber or particle mesh sizes were employed in some papers, composites with a fiber size lower than 180 μm exhibited the highest values of mechanical properties when compared to polymers reinforced with fibers sized 850−180 μm and 2360−850 μm [103]. Considering the particle size of biomass waste in the form of sawdust, some studies used particles with dimensions equal to or lower than 100 μm [69,95]. In contrast, others employed wood waste with dimensions ranging from nearly 300 to 700 μm [47,75].

Particle dimensions are a critical aspect of ensuring a successful 3D printing process for recycling seed shells. For the selected works, the average particle size ranges from 50 μm measured for cocoa shells powder [36] to the maximum size of nearly 500 μm measured for cocoa shells used as a reinforcement filler for recycled PP [52]. The plum seed shells were ground and sieved to achieve a particle size of 0.16 mm. Most works recycling fruit and seed mixed the waste with a polymer acting as a composite matrix. In a few cases, biomass waste is the main component of the extruded materials, such as in the case of 3D printing of banana peels and sugarcane bagasse using only 1% wt. of an additive like guar gum [104]. More interestingly, another work employed a biopolymer obtained from agricultural waste, i.e., hemicellulose from corn cobs, without any other modifications or blends with other polymers [50].

Regarding biomass waste derived from herbaceous plants, two types of waste pretreatments can be identified: chemical and mechanical treatment methods. On the one hand, chemical methods were mainly employed to obtain cellulose nanofibrils from sugarcane bagasse or remove hemicellulose and lignin from raw bagasse [48,86]. On the other hand, mechanical grinding was utilized to produce 100-150 μm sized flour from rice straw for thermoplastic-based composite FFF filaments, i.e., acrylonitrile butadiene styrene (ABS)-rice straw formulations [85]. Among mechanical treatments, milling is one of the commonly used methods to obtain powders, for example, from rice husks, hemp hurds, and rice husks [56,59,90], with particle sizes ranging from 1 to 200 μm. Large particles (2-25 mm) of hemp shives were also successfully printed with ceramic binders [82]. However, in most cases, the particle size range is lower than 500 μm [44,52] or even 200 μm [53,61,77] to avoid any phenomena of nozzle clogging during the extrusion. Sugarcane bagasse was also printed without any chemical treatments, only after milling and sieving, thus enabling their extrusion-based 3D printing technology without using any additional polymer matrix [98,104].

Table 3. List of the selected works analyzed according to the waste category, origin, source, type, and supplier

Waste category	Year	Ref.	Waste origin	Waste source	Waste type	Waste supplier
Wood and timber (Plant-derived)	2016	[68]	Post-industrial	Wood (Generic)	Fibers	Fibers from local industries
	2017	[57]	Post-industrial	Wood (Generic)	Flour	Flour of white softwood from local industries
	2018	[46]	Post-industrial	Spruce	Fibers	Fibers from local paper industries
		[69]	Post-industrial	Wood (Generic)	Sawdust	Furniture manufacturing companies
		[75]	Post-industrial	Pine	Sawdust	Logging into the timber industry
		[93]	Post-industrial	Wood (Generic)	Sawdust	Woodwork machines from fabrication labs
	2019	[38]	Post-industrial	Eucalyptus, Pine, Hardwood, Maple	Flour	Fibers from timber industries
		[39]	Post-industrial	Spruce	Chips	Lignin extracted from cooking liquor
		[40]	Post-industrial	Pine	Chips	Chips from a local sawmill
		[89]	Post-industrial	Teak	Fibers	Waste from local sawmills
		[103]	Post-industrial	Poplar	Fibers	Poplar fibers from the milling process
		[47]	Post-industrial	Bamboo	Sawdust	Fibers from local timber industries
		[71]	Post-industrial	Wood (Generic)	Sawdust	Woodwork machines from fabrication labs
		[49]	Post-industrial	Pine	Fibers	Fibers from local industries
	2020	[51]	Post-industrial	Wood (Generic)	Flour	Wood flour from furniture industries
		[95]	Post-industrial	Orange wood	Sawdust	Sawdust from orange pruning
		[83]	Post-consumer Post-industrial	Maple, pine	Sawdust	Sawdust from local waste
		[78]	Post-industrial	Oil palm	Fibers	Fruit brunch from local industries
	2021	[60]	Post-industrial	Wood (Generic)	Flour	Wood flour from furniture industries
		[43]	Post-industrial	Eucalyptus	Sawdust	Sawdust from the local industrial waste stream
		[84]	Post-industrial	Wood (Generic)	Fibers	Fibers from agricultural by-products

Table 3. Continues.

Waste category	Year	Ref.	Waste origin	Waste source	Waste type	Waste supplier
Fruit and seed (Plant-derived)	2017	[101]	Post-industrial	Macadamia nut	Shells	Agricultural waste
		[36]	Post-industrial	Cocoa bean	Shells	Raw and unprocessed shell waste
	2019	[81]	Post-industrial	Coffee	Grounds	Oil-extracted spent coffee grounds from supercritical oil extraction
	2020	[50]	Post-industrial	Corn	Cobs	Corn cobs from agricultural waste
		[73]	Post-industrial	Plum seed	Shells	By-products from local waste
		[95]	Post-industrial	Carob, Orange, Tomatoes	Biomass, flour, peels	Scraps from Sicilian cherry tomatoes, discarded carobs, orange peels from pruning
		[83]	Post-consumer Post-industrial	Walnut, cocoa bean, olive	Shells, pomace	Olive pomace, walnut, and cocoa shells from local waste
		[91]	Post-industrial	Soybean	Hull	By-products from local companies
	2021	[54]	Post-industrial	Cocoa bean	Shells	Cocoa bean shells from local industrial waste
		[62]	Post-consumer	Coffee	Grounds	Spent coffee grounds from local restaurants
		[87]	Post-industrial	Soybean	Hull	Soybean hull from local industries
		[88]	Post-consumer	Coconut	Fibers	Coconut fiber waste from local shops
		[64]	Post-industrial	Soybean	Hull	Soybean hull from local feeding industry
	2022	[104]	Post-consumer Post-industrial	Banana	Peel	Banana peels from the local fruit market
		[100]	Post-industrial	Corn	Cobs	Corn cobs from agricultural waste
Herbaceous plants (Plant-derived)	2017	[56]	Post-industrial	Rice	Husk	Rice husk from the local industries
	2018	[102]	Post-industrial	Hemp	Shives	Waste from local industries
		[85]	Post-industrial	Rice	Straw	Straw from local farmlands
	2019	[86]	Post-industrial	Sugarcane	Bagasse	Sugarcane bagasse from local mills
		[70]	Post-industrial	Cork	Powder	Residue from cork industries
		[59]	Post-industrial	Hemp	Hurd	Chips from local industries
		[96]	Post-industrial	Flax	Shives	By-products from local cultivations
		[48]	Post-industrial	Sugarcane	Bagasse	Crop sugarcane bagasse from industries
		[90]	Post-industrial	Rice	Husk	Husks from local agricultural industries
	2020	[76]	Post-industrial	Hemp	Stalk	Agricultural biomass from local farmers
		[82]	Post-industrial	Hemp	Shives	Hemp shives from local agricultural waste
		[94]	Post-consumer Post-industrial	Plant	Biomass (Generic)	Cellulose from tissue paper, printing paper, and plant matter
		[77]	Post-industrial	Hemp	Inflorescences, powder	Waste powder from hemp inflorescences
		[72]	Post-industrial	Rice	Husk	Rice husk ash from the local rice mills
		[95]	Post-industrial	Hemp	Leaves	By-products from agricultural waste
	2021	[97]	Post-industrial	Rice	Husk	Rice husk from local rice mills
		[52]	Post-industrial	Rice	Husk	Rice husk from local mills
		[53]	Post-consumer Post-industrial	Opuntia ficus indica, Posidonia oceanica	Cladodes, leaves	Opuntia ficus indica from local industries, Posidonia oceanica from local coasts
		[92]	Post-industrial	Miscanthus	Fibers	By-products from local industry
		[61]	Post-industrial	Buckwheat	Husk	By-products from local industry
		[79]	Post-industrial	Hemp	Biomass (Generic)	Biomass-fungi mixtures from agricultural biomass of local farmers
		[84]	Post-industrial	Hemp	Fibers	Hemp fibers from agricultural by-products
		[98]	Post-industrial	Sugarcane	Bagasse	Sugarcane bagasse from a local juice shop
		[44]	Post-industrial	Agave	Fibers	Agave fibers from a local tequila company

Table 3. Continues.

Waste category	Year	Ref.	Waste origin	Waste source	Waste type	Waste supplier
Herbaceous plants (Plant-derived) (continues)	2022	[74]	Post-industrial	Kenaf	Stalk	By-products from local industries
		[104]	Post-consumer Post-industrial	Sugarcane	Bagasse	Sugarcane bagasse from a local fruit market
		[45]	Post-industrial	Weed grass	Biomass (Generic)	Weed grass from local agricultural industries
		[55]	Post-industrial	Weed grass	Biomass (Generic)	Weed grass from local agricultural industries
		[65]	Post-industrial	Opuntia ficus indica	Cladodes	Opuntia ficus indica from local industries
		[66]	Post-industrial	Hemp	Biomass (Generic)	Biomass-fungi mixtures from agricultural biomass of local farmers
Animal (Animal-derived)	2018	[37]	Post-consumer	Mussels	Shells	Local waste stream
	2019	[80]	Post-industrial	Eels	Skin	Eel skins from the local fish landing center
		[42]	Post-industrial	Croaker	Guts	Farmed Atlantic croaker swim bladder waste
	2020	[83]	Post-consumer Post-industrial	Mussels, eggs	Shells	Mussel shells and eggshells from local waste
	2022	[67]	Post-industrial	Anchovy	Fishbone	Anchovy scraps from local market
Industrial (Plant-derived)	2018	[58]	Post-industrial	Papers	Pulp	Pulp collected from rejected press
	2019	[41]	Post-consumer	Paper	Pulp	Office printer paper waste
	2020	[94]	Post-consumer Post-industrial	Paper	Pulp	Cellulose from tissue paper, printing paper, and plant matter
	2021	[63]	Post-consumer	Paper	Pulp	Lab-collected printed office paper
	2022	[99]	Post-consumer	Meal	Biomass (Generic)	Food pre- and post-consumption waste stream of local hospital kitchen
Industrial (Animal-derived)	2022	[99]	Post-consumer	Meal	Biomass (Generic)	Food pre- and post-consumption waste stream of local hospital kitchen

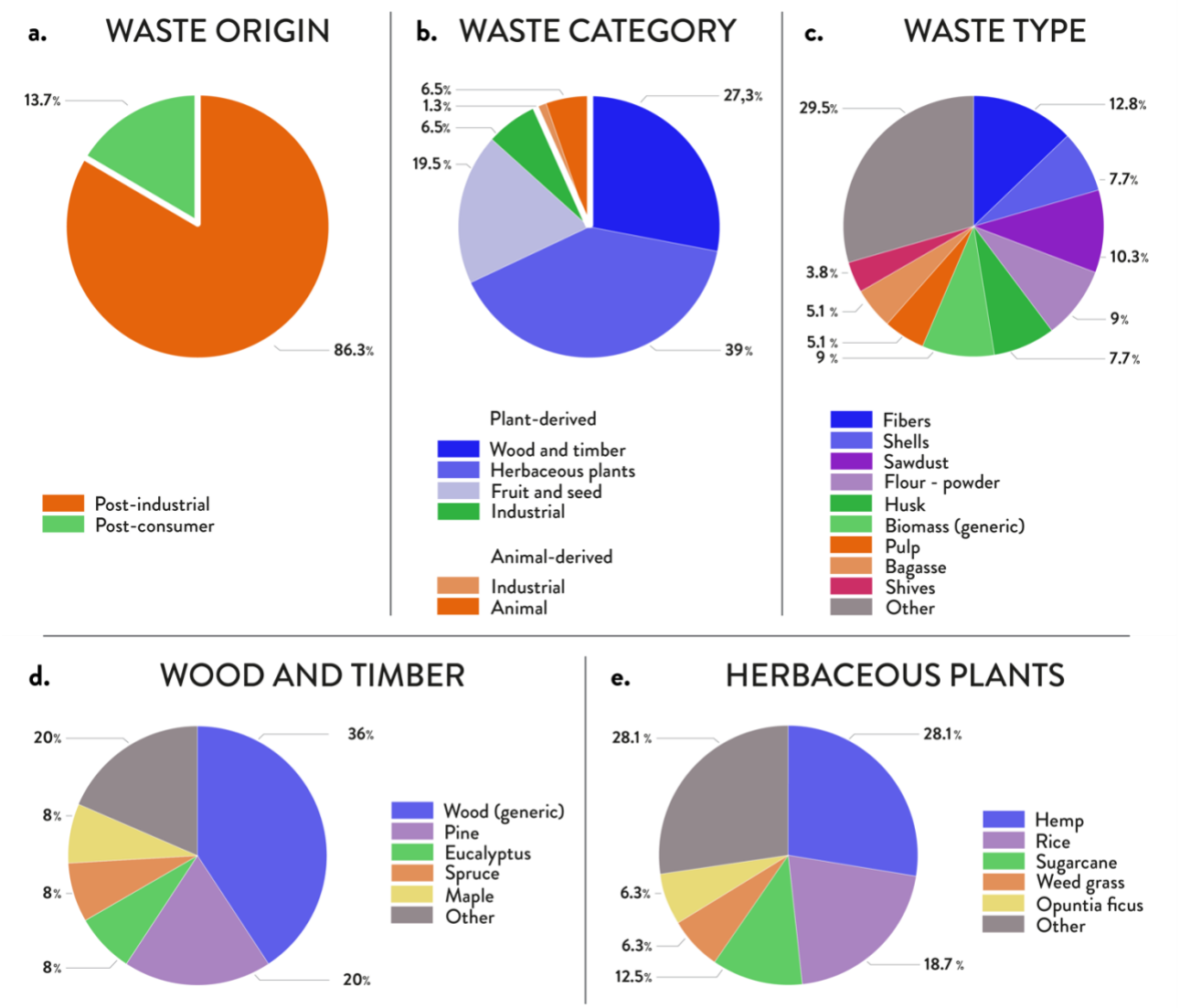


Fig. 3. Analysis of the waste: (a) origin; (b) category; (c) type; (d) insight into the wood and timber category; (e) and herbaceous plant category

3.3. New materials

This Sub-section explores the materials used with the biomass waste and scraps of the selected works. As shown in Fig. 4a and Table 4, nearly 67% of works extruded thermoplastic polymers together with biomass waste and by-products. This result is probably due to the ease of printing a thermoplastic-based filament reinforced with bio-fillers obtained with a single-screw or twin-screw extruder, as reported in many works [41,44,53,55,61,63,92]. There are also a few examples of polysaccharides used as matrix, especially for cellulose-based waste [71,86,93,94], as well as biomass powder and flour extruded either only together with water [42,50,66,79,98,99] or with water and additives, e.g., xyloglucan and guar gum, forming a sort of hydrocolloids [38,104]. Only in three cases, the matrix combined with waste derived from herbaceous plants was composed of either cement or geopolymer [72,74,82].

Among the thermoplastic materials extruded with biomass waste, PLA is the most popular thermoplastic used as a matrix in ~51% of selected studies, also considering the blend of PLA with other thermoplastics such as PLA/PBAT and PLA/PHA mixed with recycled pinewood fiber and hemp hurd powder reinforced polymers, respectively [49,59]. These thermoplastics were probably selected for their biodegradability, which is an important aspect within the framework of environmental sustainability. Apart from PLA and its blends, the other thermoplastic polymers for the 3D printing of biomass waste and scraps were PP (~10%) as well as copolyesters (CoPE, ~10%), ABS (~6%), and PE (~8%). These thermoplastic materials were probably chosen due to their low cost, easy availability, and low processing temperatures.

Considering the content of recycled biomass in the extruded materials (Fig. 4c), the most used percentages (about 70% of the papers) are in the range of 1-29% wt. [46,47,68,95]. In a few studies (~3%), a waste percentage of 40-44% was added and tested, for example, in the case of extrusion of wood waste with PLA and recycled HDPE [69,75]. However,

a higher amount can induce a detrimental effect on the mechanical properties of the wood fiber-reinforced thermoplastic [103]. A percentage of fillers obtained from bio-waste higher than 50% wt. was noticed in ~11% of selected works, which are particularly those using mussel shell powders [37,83], sugarcane bagasse powder without the use of any other additive [88], and 3D printed hemicellulose waste [50].

Table 4. List of the selected works analyzed according to the composite category, matrix, filler/reinforcement, and percentage.

Material	Year	Ref.	Matrix	Filler/reinforcement from waste/by-products	% wt.
Thermoplastic composite	2016	[68]	PLA/PHA blend	Wood fibers (shredded)	30
	2017	[101]	ABS	Macadamia nutshell fibers (shredded)	19-29
		[56]	PBAT	Silica/carbon hybrid nanoparticles	0.5-1.5
		[36]	PCL	Cocoa shell powder	10-50
	2018	[46]	BioPE	Thermomechanical pulp fibers	10-20
		[69]	PLA	Wood waste	30
		[102]	PLA	Hemp powders	1-5
		[75]	Recycled HDPE	Wood sawdust	15
		[85]	ABS	Rice straw flour	5-20
		[58]	BioPE	Thermomechanical pulp	10-30
	2019	[39]	PLA	Lignin	20-40
		[40]	PHB	Biorefinery lignin	10-50
		[89]	PLA	Teak wood fibers	1-5
		[103]	PLA	Poplar fibers	20
		[70]	TPU	Cork powder	1-5
		[47]	PLA	Bamboo fibers	10-15
		[59]	PLA/PBAT blend	Hemp hurd powder	8-27
		[96]	PLLA, PBAT, PLLA/PBS blend	Flax fibers	10-30
		[81]	PLA	Spent coffee grounds	5-20
		[48]	PLA	Sugarcane bagasse fibers	3-15
		[41]	Recycled PP	Cellulose	5-20
		[49]	PLA/PHA blend	Pinewood fibers	30
		[90]	PLA	Rice husk or wood flour	10
	2020	[51]	PLA	Wood flour	1-7
		[77]	PLA	Hemp inflorescences	10-25
		[73]	PLA/PHB blend	Cellulose nanocrystals	1
		[95]	PLA	Hemp biomass, cherry tomatoes scraps, carob flour, or orange pruning	10-25
		[91]	Co-PE	Soybean hull	5-10
		[78]	ABS	Oil palm fibers	5
	2021	[52]	Recycled PP	Rice husk (Shredded)	5-10
		[53]	PLA	Opuntia ficus indica or Posidonia oceanica	10-20
		[60]	PLA	Wood flour	5
		[92]	PTT	Miscanthus biocarbon	5-10
		[61]	PLA/PBAT blend	Buckwheat husk	5-10
		[54]	Recycled PP	Cocoa bean shell (Shredded)	5
		[43]	PLA/PBS blend	Cellulose nanofibers	1-5
		[84]	PLA	Hemp fibers, wood fibers	20-30
		[87]	CoPE	Soybean hull fibers	5-10
		[88]	PP	Coconut fibers	10-50

Table 4. Continues.

Material	Year	Ref.	Matrix	Filler/reinforcement from waste/by-products	% wt.
Thermoplastic composite (continues)		[44]	PLA	Agave fibers (Shredded)	3-10
		[63]	PLA	Paper pulp	5-15
		[64]	Recycled HDPE and recycled PP blend	Soybean hull biocarbon (pyrolyzed)	20
	2022	[45]	CoPE	Weed grass (Shredded)	5-20
		[55]	PLA	Weed grass (Shredded)	10-20
		[65]	CoPE	Opuntia ficus indica (Shredded)	10
		[67]	PLA and CoPE	Anchovy fishbone (Shredded)	10-20
Thermoset composite	2017	[57]	Urea-formaldehyde	Wood flour	13
	2021	[62]	Epoxy resin	Biochar	1-3
Ceramic composite	2020	[82]	Concrete	Hemp shives	30
		[72]	Concrete	Rice husk ash	20
	2022	[74]	Geopolymer	Shredded kenaf fibers	3
Polysaccharide composite	2018	[93]	Chitin	Cellulose (fungus-like material)	12
	2019	[86]	Alginate	Cellulose nanofibrils	20
		[80]	Alginate	Eel skin collagen	9-23
		[71]	Chitin	Cellulose (fungus-like material from [93])	9-50
	2020	[94]	Chitin	Cellulose	n.a.
		[83]	Sodium alginate	Mussel shell or eggshell powder, walnut shell or cocoa shell powder, olive pomace, pine, or maple sawdust	15-61
Hydrocolloids	2018	[37]	Sugar, water	Mussel shell powder	70
	2019	[38]	Water, Xyloglucan	Cellulose nanocrystals, wood fibers	5-30
	2021	[97]	Water, guar gum	Rice husk	42
	2022	[104]	Water, guar gum	Sugarcane bagasse and banana peels flour	32
Water-based pastes	2019	[42]	Water	Swim bladder powder	4.8
	2020	[76]	Water	Biomass, psyllium husk powder (Mycelium)	19
		[50]	Water	Hemi cellulosic paste	86
	2021	[79]	Water	Biomass, wheat flour, psyllium husk powder	18
		[98]	Water	Sugarcane bagasse powder	53
	2022	[66]	Water	Biomass, wheat flour, psyllium husk powder	18
		[99]	Water	Gelatinized starch rich phase	15
		[100]	Water	Lignin and hemi cellulosic paste	38

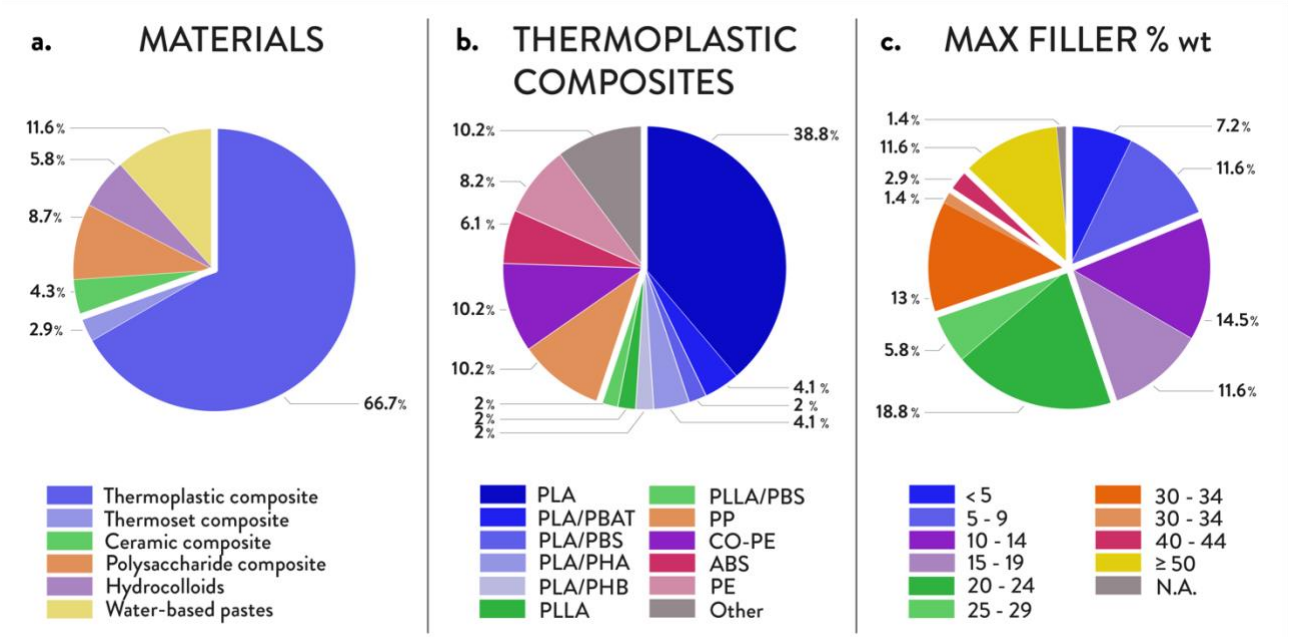


Fig. 4. Analysis of the new 3D printable materials: (a) composite category; (b) thermoplastic polymer-based composite materials; (c) weight percentages of filler/reinforcement from the waste source.

3.4. Extrusion-based AM processes

This Sub-section focuses on analyzing the extrusion-based AM processes considered from the selected works. As mentioned before, this review considers the "Material Extrusion" category of AM defined by ASTM [21], which means those processes that fabricate the final part by extruding viscous or melt materials through a nozzle. FFF (Fused Filament Fabrication, or FDM, Fused Deposition Modelling) and DIW (Direct Ink Writing, or LDM, Liquid Deposition Modeling) represent the main processes of this category. The former is commonly used with filaments made of thermoplastics or thermoplastic composites, whereas the latter allows processing liquid resins and viscous pastes at room temperature, i.e., ceramics, hydrocolloids, thermosets, and pastes. Some examples of new 3D printable materials from biomass are shown in Fig. 5: a TPU matrix filled with cork waste for FFF by Gama et al., a mussel shell-filled hydrocolloid for DIW by Sauerwein and Doubrovski, and a corn cobs hemicellulosic paste for DIW by Bahçegül et al. [37,50,70].

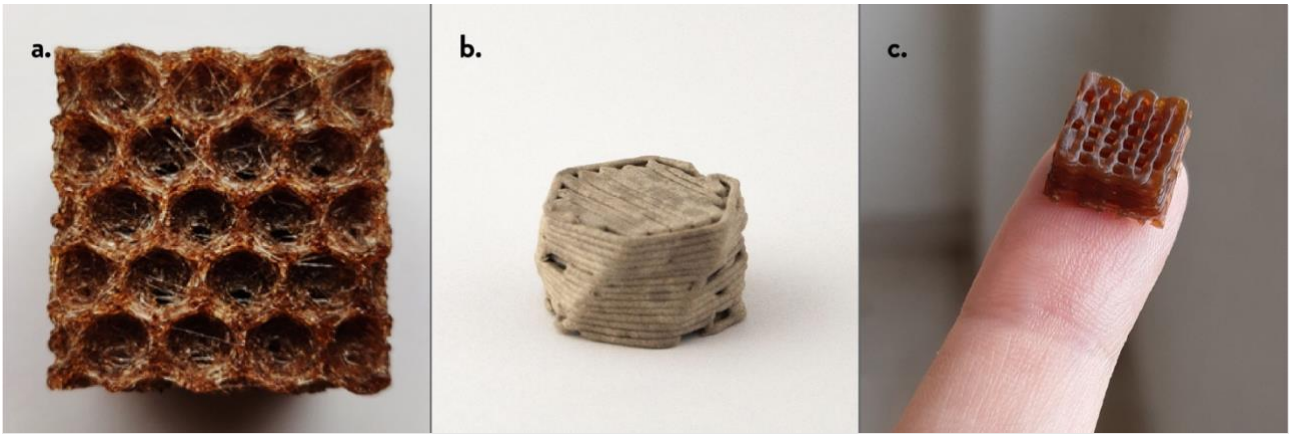


Fig. 5. 3D printed materials from biomass waste: (a) cork/TPU composite foams [70] (FFF, Reprinted with the permission of Elsevier); (b) mussel shells/sugar water composite [37] (DIW, Reprinted with the permission of Elsevier); (c) hemicellulosic paste from corn cobs [50] (DIW).

According to Table 5 and Fig. 5a, more than two-thirds of the works focus on FFF processes. This result is in line with the analysis of the new materials (Sub-section 3.3), which shows that thermoplastic composites were the most developed materials from biomass waste or by-products. Actually, FFF is a well-established group of 3D printing processes, and these 3D printers are easily accessible to a broader audience of users, ranging from low-cost desktop-size apparatus to large-scale industrial machines. Developing new materials for FFF, especially considering biomass waste and by-

products, is, therefore, more easily accessible and suitable for scaling-ups in the short- and mid-term, paving the way for the spread of new applications [19]. However, DIW can process a more comprehensive range of material categories than FFF. Consequently, it is a valid option for all those materials and biomasses that are not 3D printable by using FFF 3D printers, i.e., hydrocolloids or concrete filled with biomass [72].

As shown in Fig. 6a, about 87% of the 3D printing apparatus operates with small building volumes, confirming that new materials on small printer formats are easier to handle at the beginning of their development. Nevertheless, this consideration may not be valid for DIW processes. Processing different material categories also means tackling various potential applications and dealing with different orders of magnitude. The works focused on DIW processes show a more significant variation in the 3D printer format than FFF-related publications. Almost one-third of the DIW 3D printers are large format systems, whereas less than 10% of the FFF 3D printers are large format apparatuses. According to these results, DIW appears as a more versatile process for biomass raw materials with a broader range of applications, from small format bioprinters to large format structures for the building sector [71,80,86,94]. Moreover, small format FFF 3D printers often use filaments as primary feedstock, adding further reprocessing to obtain new 3D printable materials [31]. These steps may increase the reprocessing time, cause degradation of the raw materials, or consume more energy for melting thermoplastic materials, i.e., through further heating cycles for filament extrusion [106].

Similarly, developing custom 3D printing systems and setups is more evident for DIW processes. As from Fig. 6b, almost 25% of the publications use a custom 3D printer, either FFF or DIW system. In general, these 3D printers are not commercial solutions, and they may be developed or modified by the research team [97,98,104,107] or derived from previous works of different research groups [82,108]. From this review, DIW custom 3D printers are seven times as many FFF custom systems, representing almost two-thirds of the total DIW 3D printers. In this case, customization may be linked to lower TRLs (Technology Readiness Level) of DIW 3D printers, which initially emerged in the early 2000s [109,110]. Hence, custom systems have been developed to better control the 3D printing parameters of these new materials for DIW, defining ad-hoc solutions to process viscous pastes at room temperature, i.e., using screw extruders or pump systems on robot arms [74,83,93]. Moreover, still regarding DIW processes, no need for heating reduces the consumption of energy and increases their sustainability. Focusing on FFF, low-cost small format 3D printers were often selected for the experimentations, confirming the well-established use of this specific extrusion-based AM process, as well as its increasing interest in circular economy contexts [19,22,26,31].

These differences are also visible from the main 3D printing parameters reported in Table 5. DIW processes often mean dealing with bigger nozzle diameters, lower speeds, and thicker layers than FFF. On the one hand, these specifics result in lower 3D printing resolutions but, on the other hand, in a broader range of 3D printable materials and recovered biomass waste and by-products. Moreover, DIW processes usually do not require the use of heating during printing, minimizing the impact of high temperatures on biomass. However, the selection of temperatures during the extrusion in FFF processes was also based on the degradation temperatures of waste materials [45,47]. For these reasons, DIW represents a valuable complementary process to FFF within this context, although further development is required to foster its real exploitation.

Table 5. List of the selected works analyzed according to the extrusion-based AM process, the printer format, nozzle diameter, printing speed, layer height, and 3D printer systems and specs.

Extr-based AM	Year	Printer format	Ref.	Other specs	Nozzle (mm)	Speed (mm/s)	Layer h. (mm)	3D Printer
FDM – FFF	2016	Small	[68]	//	//	18	//	Prusa i3 Rework
	2017	Small	[101]	//	0.5	//	//	Leapfrog Creatr
		Small	[36]	//	0.6	50	0.3	Prusa i3 Hephestos
	2018	Small	[46]	//	0.4	15	//	Ultimaker Original
			[102]	//	//	//	0.1	Ultimaker 3
			[85]	//	1	//	0.2	Printrbot Simple Metal
			[58]	//	0.4	15	//	Original Prusa i3
		Large and small	[69]	//	0.5	62.5	0.15	Delta-type RepRap, Re:3D Gigabot version GB2
			[75]	Robot arm, custom	//	30	0.3	High-speed single screw extruder on a robot arm and Delta 3D printer

Table 5. Continues.

Extr-based AM	Year	Printer format	Ref.	Other specs	Nozzle (mm)	Speed (mm/s)	Layer h. (mm)	3D Printer
FDM – FFF	2019	Small	[39]	//	0.4	35	//	Prusa i3 MK3S
(continues)			[40]	//	0.75	25	0.3	MakerGear V2
			[89]	//	0.4	//	0.2	MakerBot Replicator Z18
			[70]	//	//	60	0.2	Anycubic Chiron
			[47]	//	//	20	//	low-end 3D printer after nozzle adaptation
			[59]	//	0.4	60	0.15	Da Vinci 1.0 Professional
			[96]	//	1	13-25	//	Prusa i3 Rework
			[81]	//	0.4	//	//	Einstart-S
			[48]	//	0.6	40	0.1	S1Architect
			[41]	//	0.8	20-50	0.2	Lulzbot Taz 6 MEAM
			[49]	//	0.4	23	0.2	Prusa i3
			[42]	//	0.4	30	0.2	Shenzhen 605 S model
			[90]	//	0.75	//	//	Makergear M2
		Large	[103]	//	7.62	//	5.84	Big Area Additive Manufacturing (BAAM) system at Oak Ridge National Laboratory
	2020	Small	[51]	//	0.4	//	0.1	Flythinking FS-14
			[77]	//	0.6	60	0.25	D300 Technology®
			[73]	//	0.4	33	//	WASP Delta 2040 Turbo 2
			[95]	//	0.6	//	//	D300 Technology®
			[91]	//	0.5	30	0.2	Desktop Printrbot
			[78]	//	//	//	//	UP plus 2 model
	2021	Small	[52]	//	0.8	60	0.25	Fusedformcorp 3D FF STD Doppia
			[53]	//	//	45	0.1	Sharebot Next Generation
			[60]	//	0.4	50	0.4	FS-200 printer (Guangzhou Flythinking Co)
			[92]	//	0.5	35	0.3	Lulzbot Taz 6
			[61]	//	0.8	50-80	0.15	Prusa i3 MK3
			[54]	//	0.8	60	0.25	Fusedformcorp 3D FF STD Doppia machine
			[43]	//	0.4	60	0.3	Wanhao Duplicator i3 plus
			[84]	//	//	//	0.1	Sharebot NG 2
			[87]	//	//	//	//	Prusa i3 systems
			[88]	//	//	40	0.5	Anycubic Chiron
			[44]	//	//	50	0.3	Wanhao Duplicator 4
			[63]	//	0.4	30	0.2	Shenzhen 603S model
			[64]	//	0.5	35	0.3	Lulzbot Taz 6
	2022	Small	[45]	//	//	50	0.1	Next Generation - Sharebot
			[55]	//	//	20	0.1-0.2	Next Generation - Sharebot
			[65]	//	0.4	50	0.1	Sharebot Next Generation
			[67]	//	0.4	50	0.1	Sharebot Next Generation

Table 5. Continues.

Extr-based AM	Year	Printer format	Ref.	Other specs	Nozzle (mm)	Speed (mm/s)	Layer h. (mm)	3D Printer
LDM - DIW	2017	Small	[56]	Custom	//	//	//	Hyrel system 30 M model
			[57]	//	//	15	//	Fisnar robot 7400, a 3-axis robot
	2018	Small	[37]	Custom	0.84	5	1	Modified Ultimaker 2+
		Large	[93]	Robot arm, custom	7	//	3	Six-axis articulated industrial robot with a precision material dispenser and a material pump system
	2019	Small	[38]	Custom	1.291	5	//	Modified Hyrel3D 30M with a syringe extruder
			[86]	Bioprinting	0.58	3	//	Regemat3D printing unit (version 1.0)
			[80]	Bioprinting	0.5	//	//	BIOBOT Allevi
		Large	[71]	Robot arm, custom	7	40-60	3.5-7	Six-axis articulated industrial robot with a precision material dispenser and a material pump system
	2020	Small	[76]	//	4	//	//	Delta wasp 2040 clay mode
			[50]	Custom	0.54-0.68	//	//	Custom-made 3D printer built with 3D4E
			[83]	Custom	1.6	6	1.1	Ultimaker 2+ modified for paste printing with the Stoneflower Ceramic matrix 3D Printing KIT Basic
		Large	[82]	Custom	20	//	5-10	Custom-made 3D printer extruder inspired by [108]
			[72]	//	//	12.5	//	Custom screw-type extruder mounted on a three-axis gantry system, Kenyo 3D printer
		Large and small	[94]	Robot arm, custom	1-6.5	50-150	//	Custom-made apparatus (small and large format)
	2021	Small	[97]	Food printing, custom	0.82	//	//	Custom delta (Controlled Additive-manufacturing Robotic Kit) by [107]
			[79]	//	6	30	6	Delta WASP 2040 clay (extruder adaptation)
			[62]	//	0.48	50	0.4	Hyrel-30M benchtop
			[98]	Food printing, custom	0.82	//	//	Custom delta (Controlled Additive-manufacturing Robotic Kit) by [107]
	2022	Small	[104]	Food printing, custom	1.2	7-12	//	Custom delta (Controlled Additive-manufacturing Robotic Kit) by [107]
			[66]	Custom	6	30	6	Delta WASP 2040 clay (extruder adaptation)
			[99]	Food printing	0.8	30	//	Foodini (Natural Machines)
			[100]	Custom	0.54-0.68	//	//	Custom-made 3D printer built with 3D4E
		Large	[74]	Custom	11-28	//	//	Self-made apparatus with different nozzle sections

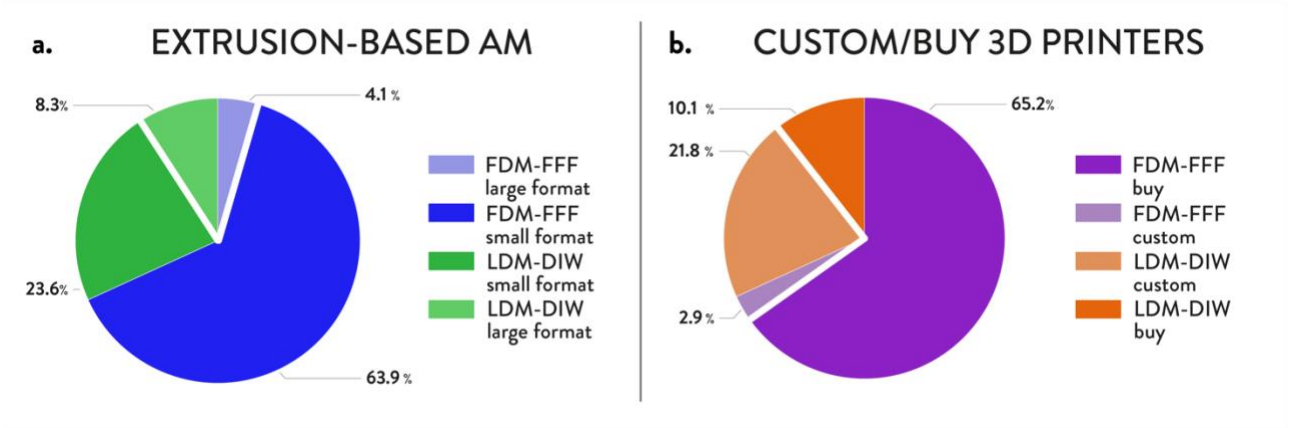


Fig. 6. Analysis of the extrusion-based AM processes: (a) process and printer format; (b) custom/buy 3d printer systems.

3.5. Applications and case studies

This Sub-section aims to depict the emerging applications and trends related to new 3D printable materials from biomass waste and by-products. According to the general analysis of Sub-section 3.1, more than 50% of the works analyzed in this review are linked to materials science-related research areas. In general, these works mainly focus on the characterization of the new materials from biomass rather than defining a possible field of application through a practical case study. For this reason, some applications from the design practice and industrial sectors were also selected to highlight possible trends for using these new materials, showing some possible ways of exploitation.

3.5.1 Applications from the literature review

The selected works were analyzed to understand whether the experimental activities were linked to possible fields of application or practical demonstrations. As from Fig. 7a, about half of the publications include some hypothesis on possible applications or potential uses (~51%), but only one-third of the works show a real case study or a practical demonstration with the new materials through 3D printed products or prototypes. In this case, 3D printing was not only used for printability assessment and material characterization but also to demonstrate the potential use of 3D printable materials within the real context. The specific products, applications, and general fields are visible in Table 6. As from the previous Sub-section, more applications are linked with the FFF process (14 works), confirming its prevalent use compared to DIW systems (8 works). Despite the limited number of applications, seven main fields were defined to cluster the publications with a specific application or demonstration ("buildings and architecture", "furniture", "equipment", "accessories", "prosthetics and medical", "packaging" and "bioengineering"). As shown in Fig. 7b, the most represented fields of application are "buildings and architecture", "furniture", and "equipment", more than 50% of the applications. In this case, different products were 3D printed as proofs-of-concept, ranging from small furnishing accessories, i.e., drawer knobs and lamps [69,83], to large architectural structures, i.e., sculptures (Fig. 8a) and turbine blades [71,94], dealing with both FFF and DIW. More technical fields of application are mainly related to one of the two extrusion-based AM processes, such as "bioengineering" for scaffolds production (DIW) [80] or "prosthetics and medical" focused on customized orthoses (FFF) [95].

Table 6. List of the selected works analyzed according to the application sector and the specific product or application.

Year	Extr-based AM	Ref.	Area	Product/Application
2018	FDM - FFF	[69]	Furniture	Desk cable feedthrough parts, drawer knob
	LDM - DIW	[37]	Equipment	Soil fertilizer flowerpot
		[93]	Buildings, architecture	Wind blade turbine
2019	FDM - FFF	[39]	Accessories	Smartphone protective case
		[103]	Furniture	Podium base
		[47]	Equipment	Brake levers of a bike
		[49]	Equipment	Technical part for 3D printing
		[42]	Other	Moisture actuators for meteor-sensitive architectures, biomedical devices, soft robotics
	LDM - DIW	[38]	Buildings, architecture	Mashrabiya, a wooden window (Patent application)
		[80]	Bioengineering	Scaffold for tissue fabrication
		[71]	Buildings, architecture	Wind turbine blade
2020	FDM - FFF	[51]	Furniture	Scaled chair model
		[94]	Buildings, architecture	Sculpture (Hydra)
		[77]	Prosthetics, medical	Neck orthosis
		[95]	Prosthetics, medical	Customized neck orthosis and laryngoscope
	LDM - DIW	[50]	Bioengineering	Scaffold prototype for biomedical application
		[83]	Furniture, accessories	Lamp, Hairpin
2021	FDM - FFF	[43]	Packaging	Fruit crate and compostable packaging
		[84]	Buildings, architecture	Geogrids for Construction and Demolition sector
		[64]	Equipment, accessories	Shaft and elliptical gears, spectacle frames
	LDM - DIW	[98]	Packaging	Customized food package casings
2022	FDM - FFF	[65]	Other	Controlled release fertilizers for agriculture

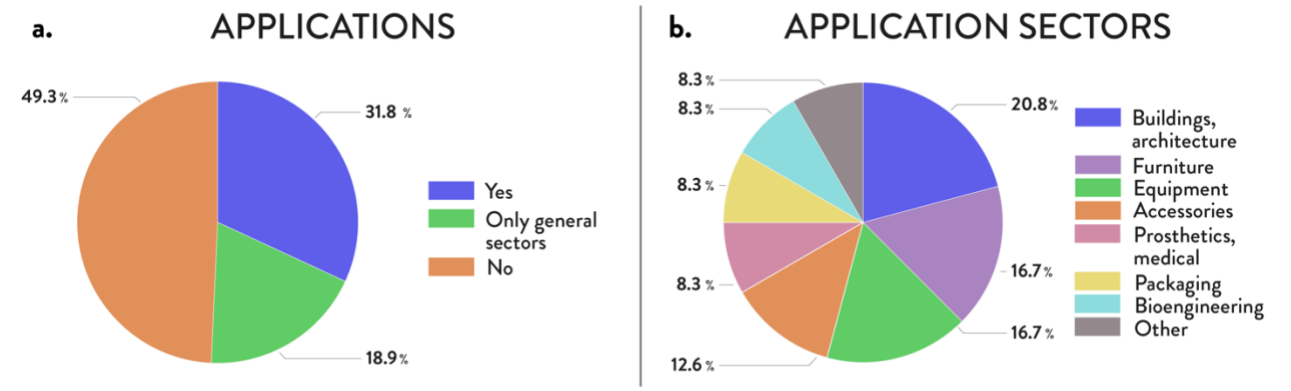


Fig. 7. Analysis of the applications from the selected works: (a) development of applications; (b) main application sectors.

In the perspective of real circular bioeconomy practices, biodegradability and even compostability represent crucial features for these products, especially according to the application field. On the one hand, durability should be ensured in long-lasting applications during the whole lifecycle of a specific product, resisting biodegradation before its End-of-Life, i.e., buildings, architecture, and furniture. On the other hand, some sectors aim to develop new products with a shorter lifecycle, such as for packaging products or temporary orthoses, which should be easily and rapidly decomposed by microorganisms after disposal [111,112]. In both cases, the focus is on developing new products that can be decomposed without any environmental pollution if not properly reused or recycled. However, among the selected studies dealing with applications, only one work focused on these aspects. The study assessed the biodegradability of 3D printed food package casings from sugarcane bagasse, showing a weight decrease of almost 50% after burying in soil for five weeks due to the microbial action [98]. Hopefully, these types of tests will be incremented in the future to prove the real circular exploitation of agricultural waste in additive manufactured products and help in designing new products according to their expected lifecycle.

Notwithstanding the potential biodegradability and the consequent limitation of robustness for these products, nearly 75% of selected works linked to applications studied the mechanical properties of their 3D printed materials, and some of them observed elastic modulus and tensile strength values (1-2 GPa and 20-45 MPa, respectively) comparable to commodity polymers [39, 43, 47, 51, 77, 95]. In specific cases, which showed for example 3D printed parts made of PLA-based composites filled with hemp waste for biomedical applications, higher elastic modulus values of about 3-4 GPa were found [77, 95]. Two studies also presented 3D printed objects made of recycled wood waste with mechanical properties of natural woods such as medium to low-density woods and high-density foams, with potential applications in architecture [38] and energy sectors for the fabrication of wind turbine blades [93]. However, low values of strain at break (< 10%) were reported in the works mentioned before, indicating the need for improved tenacity, especially for demonstrators with consequences to people safety. In summary, these results demonstrate the potential of 3D printable materials from agro-waste in different applications, underlining the need for a correct matching between the materials' properties and the new application to be developed.

3.5.2 Commercial case studies

The selected best practices from the design practice and industrial sectors confirm the increasing interest in using biomass waste and by-products as the primary source for new 3D printable materials. Three main trends may be detected: production and selling of commercial materials; production and sale of commercial materials and 3D printed products; design and production of 3D printed products. In the first case, different stakeholders, including designers and end-users, can easily buy new filaments for FFF derived from biomass, such as Woodfill and Corkfill from Colorfabb BV [113]. Other industrial realities allow not only to buy commercial material from their site but also to order some on-demand 3D printed products from their e-commerce. For example, “Hemprinted” is an online shop that sells filaments and furnishing products made from agro-waste, such as hemp biomass (Kanèsis filaments, Fig. 8b) [114]. In addition, emerging design studios and professionals are developing new products from biomass materials, following the Material Driven Design (MDD) approach [115]. The design studio “Krill Design” has been developing new filaments for FFF in collaboration with agri-food industries starting from their biomass waste and by-products, i.e., orange and lemon peels. Furthermore, it is using these materials for new products such as “Ohmie – the Orange Lamp” (Fig. 8c) [116].

These exemplary best practices show possible synergies between different industry stakeholders to create new circular economy strategies from upcycling biomass waste, fostering the real exploitation of these materials. Some connections between academic research and practical contexts may be possible by promoting the knowledge of these new materials amongst design practitioners and industrial realities for their use in real applications [19]. This aspect will address the

typical challenge of knowledge integration and technology transfer from academia to industrial partners, as identified for any new processes in bioeconomy [117]. However, there are still other challenges to be overcome to make AM processes truly successful innovative technologies within the bioeconomy, such as more widespread adoption of AM technologies in business-to-business (B2B) context and an increased cost competitiveness of bio-based waste 3D printed products with respect to fossil-based and bio-based ones.

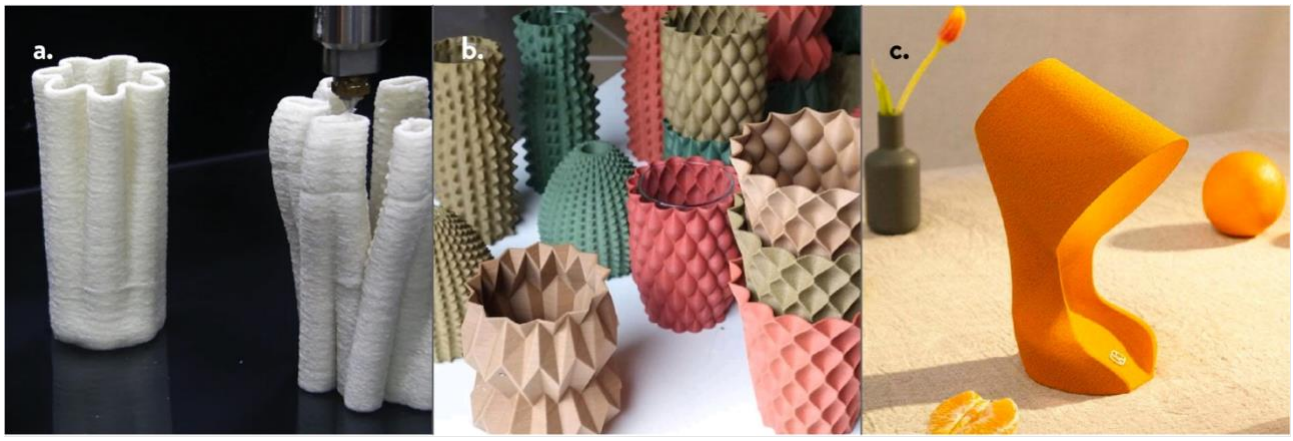


Fig. 8. Examples of 3D printed applications from biomass waste: (a) "Natural Composite Pillar" made of chitin/cellulose composite through large format DIW [94]; (b) "Hemprinted" e-shop of products and filaments for FFF made of PLA filled with hemp, tomato, and pomegranate waste [114]; (c) "Ohmie Lamp" made of PHB filled with orange peels through FFF by Krill Design [116].

3.6. Limitations and future perspectives of waste biomass materials for extrusion-based AM processes

The main limitations and future perspectives are resumed in this section, aiming to stimulate further investigation and practical exploitations of these emerging materials for extrusion-based AM processes.

Focusing on the limitations, there are some issues and drawbacks to overcome to foster the use of biomass waste and extrusion-based AM in real contexts. In detail:

- Mechanical treatments performed to obtain dimensions suitable for 3D printing, e.g., grinding of bio-based waste, require energy consumption and have to be optimized in terms of energy expenditure.
- Variability of the biomass waste may negatively affect the 3D printing process, increasing the failure rates, i.e., nozzle clogging caused by different granulometries of the biomass particles.
- The use of high temperatures during the FFF printing processes is detrimental from an environmental point of view, and it may also damage biomass fillers or reinforcements.
- The development of some extrusion-based AM processes is still ongoing, as well as some materials from biomass waste, requiring additional work in the short time for their implementation, as well as further resources and time.
- The limited cost competitiveness of bio-based waste 3D printed products compared to fossil-based and bio-based materials may lead to the latter solutions for new applications in the short term.

Some promising aspects can also be detected according to the previous analysis. These strengths points can be seen as current beneficial aspects and show some future perspectives and opportunities to work on. For instance:

- Biomass waste and scraps are obtained from industrial activities in most of the analyzed works, paving the way for adopting bio-waste as input materials for AM processes in B2B contexts.
- Extrusion-based AM can reprocess a wide variety of biomass, i.e., different waste categories and types.
- The biodegradability of the most thermoplastic materials used as matrices for bio-waste reduces the negative effects in case of uncontrolled landfilling of 3D printed products with bio-waste.
- In view of reducing energy consumption, the absence of heating for DIW processes increases the sustainability of the processes, contributing to lower impacts.
- The flexibility of AM allows designing new applications from small to large format products, as well as to be implemented in different application fields, i.e., prosthetics, bioengineering, furnishing, and architecture.
- The development and commercialization of some applications are gradually spreading, showing the interest from industry stakeholders, designers, and professionals in implementing these aspects in their current and future projects,

Although further work should be done to improve extrusion-based AM processes for biomass waste, this technology appears as a promising way to implement the principles of circular economy and bioeconomy in real contexts.

Specifically, DIW processes show the potential to allow this transition as soon as their effective scaling up and implementation in design and industrial scenarios is effectively achieved, i.e., in the mid-term.

4. Conclusions

This review systematically collected the works dealing with developing new materials from biomass waste or by-products for extrusion-based AM processes. The main aim was to analyze the current situation in the academic field, considering these topics linked with the concepts of bioeconomy and the circular economy, focusing on the raw materials, the new 3D printable materials, the 3D printing specifics, and the possible applications. As a result, 69 works from 2016 were selected by following the PRISMA statement, defining the emerging framework with the help of selected best practices from industrial context and design practice.

An increasing interest in these topics emerges from the general analysis, especially considering the timeframe from 2019 to 2022 (56 works). Despite the interdisciplinarity of these topics, the publications are mainly linked to material science-related research areas. In addition, few works included environmental and social considerations of their work. Further work should be done to implement different perspectives on this topic, i.e., with a holistic approach that considers the environmental, social, and economic aspects of sustainability.

Most of the biomass waste and by-products recycled are derived from industrial activities like those from paper and furniture industries as well as agricultural activities. Based on this consideration, it is not surprising that timber, wood, and herbaceous plants are the most common biomass waste exploited as input for the 3D printing processes. Moreover, the size and the shape of the bio-derived waste were also identified as crucial for extrusion-based 3D printing to avoid nozzle clogging and minimize defects in the final 3D printed objects. For this reason, mechanical grinding of biomass waste was carried out in numerous works before preparing the 3D printable materials. Except for some cases, the final dimensions of bio-derived and recycled fillers were lower than some hundreds of μm.

Concerning the materials used as a matrix for bio-waste reinforced composites, the most employed polymers are PLA and its blends, for instance, PLA/PHA and PLA/PBAT, probably selected for their popularity in FF 3D printing and their biodegradability. Other thermoplastics such as PP, ABS, and copolyesters were also used to a certain extent, probably due to their quite widespread use as 3D printable materials, low costs, and good mechanical properties. Several works combined bio-waste with polysaccharides, like alginate and chitin, as well as water and other additives, due to their biodegradability and compatibility with biomass. Regarding the amount of bio-waste added to the matrix, the most common percentages are in the range of 1-29% wt., also because a higher filler content could negatively affect the final properties of the extruded materials.

The most consolidated extrusion-based AM process is FFF, and DIW can be defined as the most versatile one. The former represents a well-established and accessible solution, especially for small format low-cost 3D printers. Despite its lower TRL, the latter is used to process a wider range of materials at different scales, encouraging the development and customization of new 3D printer systems. The two processes could be seen as complementary solutions to enlarge the variety of materials from biomasses.

More than half of the works report at least some hypothetical sectors for exploiting these new materials. However, real products or demonstrations developed during the experimental activities are still limited within the selected publications. The industrial sectors and the design practice show an increasing interest in using biomass waste and by-products through AM, ranging from producing commercial filaments to designing new products. However, future research should strengthen their real exploitation, i.e., fostering the knowledge of these emerging materials amongst the design professionals and interested stakeholders, or focusing on the biodegradability and durability aspects. To conclude, this review aims to stimulate future research and projects by encouraging new interdisciplinary synergies between the academic and practical contexts to exploit new circular solutions from biomasses within the real world. The transition to a more sustainable circular bioeconomy needs to embrace the concept of biocircularity, which can avoid the trap of mimicking the conventional linear economic system of take, make, consume, dispose, even with bioeconomy [11]. The valorization of agro-waste as secondary resources to produce goods is only one of the attributes necessary for the development of biocircularity. However, AM technologies can positively affect this transition by introducing biomass waste materials as fillers in bio-based matrices (e.g., PLA) to obtain fully biodegradable products. This fact implies that the use of thermoplastic materials, i.e., PLA and its blend with other thermoplastics, represent the most widespread choice in most works analyzed in this review.

CRediT author statement

Alessia Romani: Conceptualization, Data curation, Formal analysis, Investigation, Methodology, Resources, Validation, Visualization, Writing – original draft, Writing – review & editing. **Raffaella Suriano:** Conceptualization, Funding acquisition, Project administration, Validation, Writing – original draft, Writing – review & editing. **Marinella Levi:** Funding acquisition, Project administration, Writing – review & editing, Supervision.

Acknowledgments

This research did not receive any specific grant from funding agencies in the public, commercial, or not-for-profit sectors. This review was not registered.

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