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Biomass recycling 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 analysis of the current situation was carried out on 64 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 prominent development of thermoplastic reinforced materials, i.e., PLA-filled composites, a wide range of biomass waste and/or by-products have been studied, especially for small format low-cost Fused Filament Fabrication. Although the academic field may be less focused on the exploitation of 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 model of economic development called circular economy (CE) has gained ever-increasing attention due to the need for minimizing the environmental impacts of human activities and its potential to promote well-being and harmonious society [1]. CE provides a novel concept of economic systems, promoted by several national governments around the world, aiming to maintain extraction rates of resources and preserve their value as long as possible within the system [2]. The CE concept was proposed 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 what is usually considered waste is maintained as long as possible, considering it as a resource to be reused and reintroduced into the system. Waste management plays a key 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].

To boost the transition towards the CE, it is undeniable that a strong development of the so-called bioeconomy can be beneficial, especially in dealing with materials [10]. Bioeconomy indeed aims at using renewable biological resources from biomass rather than fossil- and mineral-based resources to produce goods, foods, and energy [11]. 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 [12]. Among these, some approaches can be identified as very popular such as the incorporation of functionalized lignin in polymeric materials [13], several biological and thermochemical treatments of bio-based waste coming from domestic, agricultural, and industrial activities [14], and the production of functional bioplastics and advanced materials from agri-food waste [15].

Within this framework, additive manufacturing (AM) stands out as being one of the most used technologies to foster new circular economy models [16–18]. According to the international standard (ISO), AM is defined as a class of processes able to obtain objects starting from 3D model data, through a layer-by-layer approach, differently from subtractive manufacturing and formative manufacturing methods [19]. Additive technologies or 3D printing represent nowadays an important innovation in different sectors, offering several advantages with respect 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 [20]. For these reasons, it is considered the key to the fourth industrial revolution, based on intelligent automation technologies and smart factories [21]. Moreover, the role of AM in promoting a more sustainable economic system is emerging, considering its significant benefits, and increasing applications [22]. 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 [23], thermoplastic polymers [24], materials from the construction sector, and rubber tires [16].

Additive technologies mainly involved in a CE perspective are the "Material Extrusion" technologies or extrusion-based AM, thanks to their accessibility and flexibility [17,19]. 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 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 [25–28], while the latter the recycling of fiber-reinforced polymers and ceramics [29,30]. However, how the extrusion-based AM technologies have been employed so far for the recycling of bio-based waste is still unclear.

This work 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/or closed-loops of biomass as raw materials?"
- RQ2: "Which are the kinds of biomass used as raw materials, the new 3D printable materials, and the main characteristics of the extrusion-based AM processes related to this research context?"
- RQ3: "What are the emerging applications and trends within this research context?"

After explaining the methodology, a general analysis of 64 works from 2016 is depicted to give a frame of 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 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 the biomass waste to the potential applications, as well as its pragmatic approach to the topic.

Nomenclature

ABS	Acrylonitrile Butadiene Styrene	PBS	Polybutylene Succinate
AM	Additive Manufacturing	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
LCA	Life Cycle Assessment	PTT	Polytrimethylene Terephthalate
LDM	Liquid Deposition Modeling	TPU	Thermoplastic Polyurethane
PBAT	Polybutylene Adipate	TRL	Technology Readiness Level

2. Materials and Methods

This paper aims to analyze the works that use biomass waste and/or by-products as a starting point for new 3D printable materials through a systematic literature review, following the PRISMA systematic review statement [31,32]. The eligibility criteria are resumed in the first column of 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) [19], excluding reviews, meta-analyses, or works with a theoretical perspective on 3D printing. After a preliminary screening of the results, a timeframe from 2015 to 2022 was set since no works were found before 2016. Only works including experimental activities focused on biomass waste, scraps, and/or by-products were considered, avoiding articles dealing 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 July 2022, as reported in Table 1. Similar search strategies were followed starting from a set of common keywords, defining the two specific query strings reported in the third column of Table 1. These query strings can also resume the search strategy within the two databases, including keywords related to AM, biomass origin and waste type, and circular economy.

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/or by-products. • Works focused on Material Extrusion AM (or extrusion-based AM) [19]. • 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")
	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 screened according to the main language of the work by considering publications in English, or avoiding duplicates, retired or not accessible works, i.e., full-text not available for download. The screening was carried out starting from titles, abstracts, and keywords from one reviewer, selecting the excluded and the 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 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. One reviewer screened each full-text article, and discrepancies were resolved with the help of a second or third reviewer, reducing the risk of errors or biases. In the end, the eligibility of each record was assessed, defining an intermediate set of screened works. Two additional rounds of screening were undertaken by carefully reading the full text and assessing the data completeness. As for the previous screening, uncertainties were fixed by discussing with a different reviewer, or by searching or deducting the missing information, i.e., 3D printer format or waste origin.

Data extraction was performed by filling the previous matrix and adding new data columns related to the biomass, new material, AM process, and potential applications. Data were extracted by one reviewer, whereas another reviewer checked the matrix. Data were collected from the full text, the link of the publication, and, in case of discrepancies, from other 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 by using excel, creating a matrix to analyze specific aspects, i.e., type of waste. This matrix was also used to plot the graphs to visualize the results of the analysis and it is visible at: https://github.com/piuLAB-official/Dataset_A.Romani_2022_JCP.

Some best practices 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 by searching the commercial solutions used in the experimental activities of the screened works of this review.

3. Results and Discussion

This section presents the analysis of the 64 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 the analysis of 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.

At first, 1185 records were found in the search databases. After the preliminary check (duplicates, retired and non-English works), 1071 records were screened. The second screening included 679 reports, which were further screened, identifying 239 reports to assess for eligibility. As from Fig. 1, these two screenings significantly reduced the selected records, since 64 studies were selected to be included in this work after the review and the data extraction. Many works cited 3D printing technologies as possible future development without performing experimentations on it, or only mentioned the possibility to use biomass waste and/or by-products as raw material. Furthermore, some works focused on different research areas linked to AM, bioeconomy, and/or circular economy, i.e., Industry 4.0-related topics.

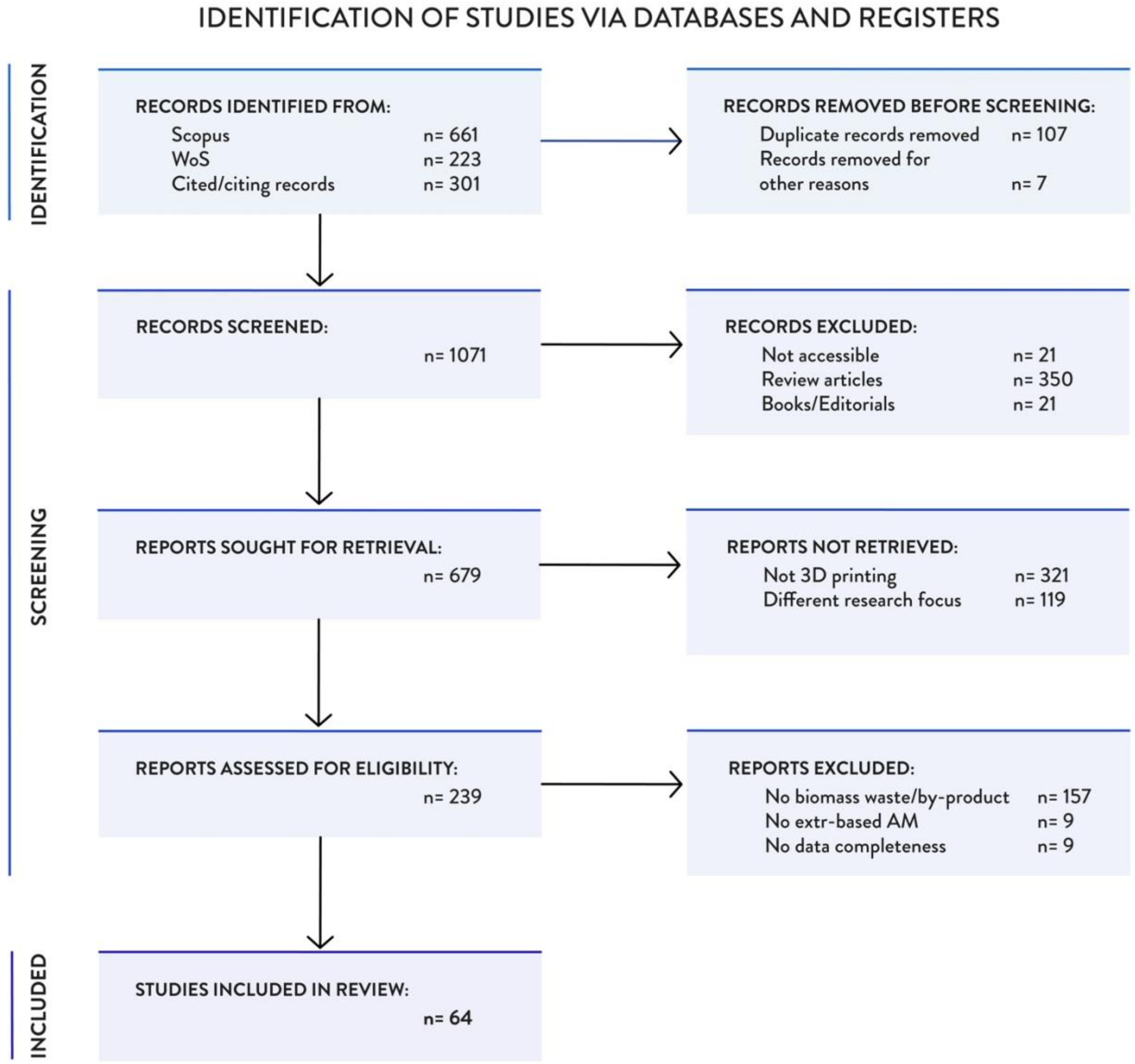

Fig. 1. PRISMA flow diagram showing the selection process of the literature review presented in this work.

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/or by-products as main raw materials. As previously seen in Fig. 1, 64 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 be probably 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 51 works, nearly 80% of the results, were found from 2019 to 2022. Furthermore, the publications of 2022 (5) may increase considering the provisional number retrieved by this review. New works could be published in the next months, and other publications may be not indexed within Scopus and/or WoS, yet.

The publications were then analyzed considering their main research area, which was determined according to the keywords and the publication source of each work. In general, only one conference paper has been found, showing that publications in academic journals are the most common way to disseminate these kinds of results. Moreover, journal articles are more frequently indexed in academic repositories rather than conference papers, hence some works could be omitted by this review for this specific reason.

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

Year	N. Ref.	References
2016	1	Le Duigou et al. [33]
2017	4	Girdis et al. [34], Biswas et al. [35], Pitt et al. [36], Tran et al. [37]
2018	8	Sauerwein and Doubrovski [38], Filgueira et al. [39], Pringle et al. [40], Coppola et al. [41], Horta et al. [42], Sanandiya et al. [43], Osman and Atia [44], Tarrés et al. [45]
2019	18	Kam et al. [46], Tanase-Opedal et al. [47], Chinga-Carrasco et al. [48], Vaidya et al. [49], Govindharaj et al. [50], Petchwattana et al. [51], Zhao et al. [52], Gama et al. [53], Depuydt et al. [54], Xiao et al. [55], Badouard et al. [56], Chang et al. [57], Liu et al. [58], Vijay et al. [59], Zander et al. [60], Guessasma et al. [61], Li et al. [62], Le Guen et al. [63]
2020	12	Bhardwaj et al. [64], Bahçegül et al. [65], Sinka et al. [66], Yang et al. [67], Sanandiya et al. [68], Calì et al. [69], Muthukrishnan et al. [70], Frone et al. [71], Calì et al. [72], Sauerwein et al. [73], Balla et al. [74], Ahmad et al. [75]
2021	16	Nida et al. [76], Morales et al. [77], Scaffaro et al. [78], Yang et al. [79], Diederichs et al. [80], Andrzejewski et al. [81], Bhardwaj et al. [82], Morales et al. [83], John et al. [84], Cislaghi et al. [85], Alhelal et al. [86], Balla et al. [87], Nida et al. [88], Gama et al. [89], Figueroa-Velarde et al. [90], Tao et al. [91]
2022	4	Kong et al. [92], Nida et al. [93], Scaffaro et al. [94], Scaffaro et al. [95], Scaffaro et al. [96]

As from Table 3, 11 different clusters were detected from the analysis of the 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 50% of the publications considered within this review, which means 36 works (Fig. 2b). Moreover, 7 works show that other sub-fields were related to materials science because the experimental activities were linked to some applications and/or practical demonstrations of different research or application fields, i.e., architecture and product design. The remaining 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 ~14%, ~12%, and ~11% 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. Therefore, materials science appears as the most involved research area at the intersection of the topics of this review although there is an increasing interest from other disciplines, fostering collaborations and multidisciplinary projects.

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]. Consequently, the publications were also reviewed to check whether they considered the environmental and social aspects through specific methodologies or experimental procedures. Only one publication includes 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 [38]. 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/or 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.

Table 3. List and number of the selected works according to the main research area.

Research area	N. Ref.	References per year
Materials science and engineering	10	**2016:** //; **2017:** [37]; **2018:** [38]; **2019:** [46], [47], [49], [60], [62]; **2020:** //; **2021:** [84], [90]; **2022:** [94].
Polymers science and engineering	10	**2016:** //; **2017:** //; **2018:** [39]; **2019:** [54], [58], [61]; **2020:** [65], [67]; **2021:** [77], [78], [83]; **2022:** [95].
Composite engineering	9	**2016:** //; **2017:** [35], [36]; **2018:** [45]; **2019:** [55]; **2020:** //; **2021:** [79], [81], [86], [91]; **2022:** [96].
Materials science for specific sectors	7	**2016:** [33]; **2017:** //; **2018:** [40] ; **2019:** [53], [59]; **2020:** [70], [71]; **2021:** //; **2022:** [92].
Manufacturing and processing engineering	5	**2016:** //; **2017:** //; **2018:** [42]; **2019:** //; **2020:** [64], [69], [75]; **2021:** [82]; **2022:** //.
Environmental sciences	5	**2016:** //; **2017:** //; **2018:** //; **2019:** [50], [57]; **2020:** [66], [73]; **2021:** [85]; **2022:** //.
Additive manufacturing	4	**2016:** //; **2017:** //; **2018:** [44]; **2019:** [48]; **2020:** //; **2021:** [87], [89]; **2022:** //.
Chemistry & Physics	4	**2016:** //; **2017:** //; **2018:** //; **2019:** [51], [63]; **2020:** [74]; **2021:** [80]; **2022:** //.
Applied sciences	3	**2016:** //; **2017:** //; **2018:** [43]; **2019:** //; **2020:** [68], [72]; **2021:** //; **2022:** //
Waste management	3	**2016:** //; **2017:** //; **2018:** //; **2019:** [56]; **2020:** //; **2021:** [76], [88]; **2022:** //
Other	4	**2016:** //; **2017:** [34]; **2018:** [41]; **2019:** [52]; **2020:** //; **2021:** //; **2022:** [93]

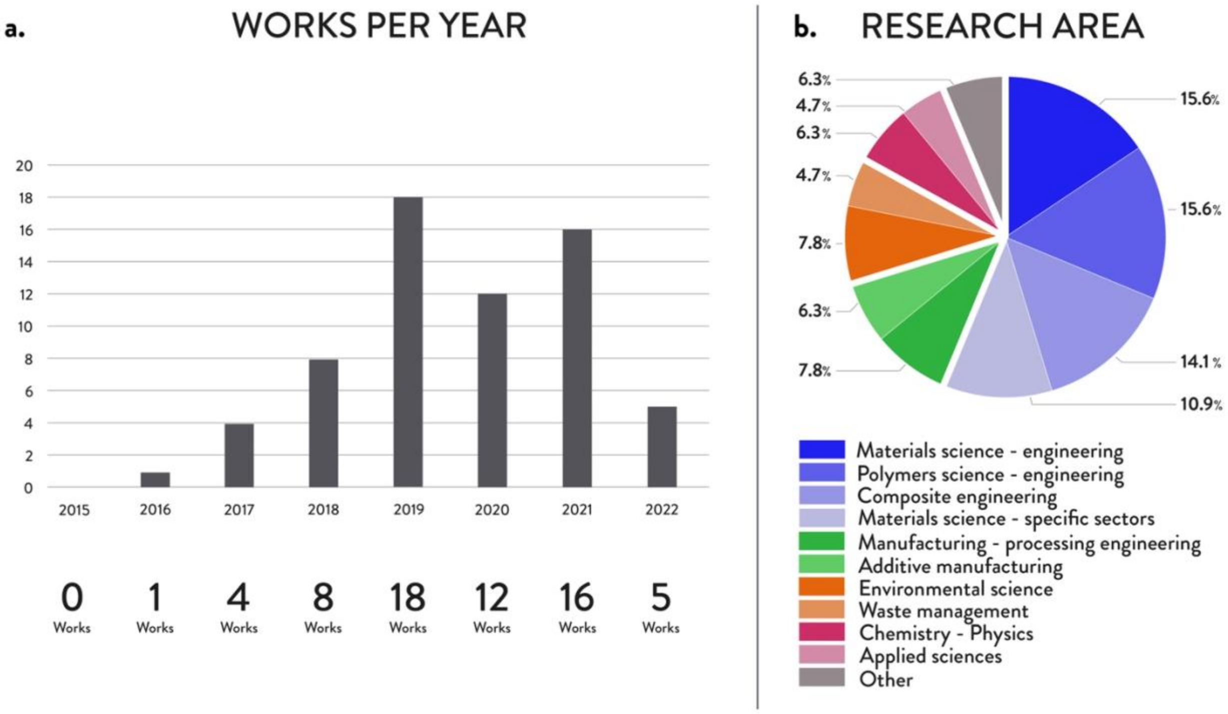

Fig. 2. General analysis: (a) works per year; (b) main research areas of the selected works.

3.2. Waste, scraps, by-products

This Sub-section focuses on the analysis of 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 and the waste originated from end-users and consumers only for nearly 16% of works. According to Fig. 3b, more than one-third of the works exploited wood and timber as waste material, which were mainly supplied as a feedstock in the form of fibers and sawdust. Still considering the wood waste type, either wood flour [36,67,72] or wood chips [47,49] were provided as feedstocks only in a few cases (Table 4). In more than one-third of the works, no explicit indications were found for the wood classification. When specified, 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 (~36%) have employed waste derived from the processing of herbaceous plants (Fig. 3c). Within the waste category of herbaceous plants, hemp was the most used as recycled feedstock in various forms for example shives [41,55,66]. Fig. 3e also shows that sugarcane in the form bagasse [48] and rice husks [77,88] were also employed as a recycled herbaceous material. The rest of the works recycled waste and by-products of fruits and seeds (more than 20%) and paper pulp, fish, and mussels (~12%).

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 [97]. To improve the reinforcement dispersion in the extruded materials, natural fibers were usually ground and sieved to obtain 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 [51] 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 [36], while dimensions lower than 75 μm were also presented by Kam et al. for DIW technology [46]. In other cases, thermomechanical pulp fibers were milled to achieve an average fiber length of 0.4 mm and a diameter of 38 μm [39]. 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 [52]. Considering the particle size of biomass waste in form of sawdust, some studies used particles with dimensions either equal to or lower than 100 μm [40,72], while others employed wood waste with dimensions ranging from nearly 300 to 700 μm [42,54].

Particle dimension is a critical aspect to ensure a successful 3D printing process also for the recycling of seed shells. For the selected works, the average particle size ranges from 50 μm measured for cocoa shells powder [37] to the maximum size of nearly 500 μm measured for cocoa shells used as a reinforcement filler for recycled PP [77]. The plum seed shells were ground and sieved to achieve a particle size of 0.16 mm. The vast majority of 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 [93]. More interestingly, another work employed a biopolymer obtained from agricultural waste, i.e., hemicellulose from corn cobs without any other modifications and blends with other polymers [65].

Regarding biomass waste derived from herbaceous plants, two different types of waste pretreatments can be identified: chemical treatments and mechanical treatment methods. On the one hand, chemical methods were mostly employed to obtain cellulose nanofibrils from sugarcane bagasse or remove hemicellulose and lignin from raw bagasse [48,58]. 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 [44]. Among mechanical treatments, milling is one of the commonly used methods to obtain powders, for example, from rice husk as well as from hemp hurds and rice husks [35,55,63], both with a particle size ranging from 1 to 200 µm. Very large particles (2-25 mm) of hemp shives were also successfully printed with ceramic binders [66]. However, in most cases, the particle size range is lower either than 500 µm [77,90] or even 200 µm [69,78,81] to avoid any phenomena of nozzle clogging during the extrusion. Sugarcane bagasse was also printed without performing any chemical treatments, only after milling and sieving, thus enabling the extrusion-based 3D printing technology of this agricultural waste without the use of any additional polymer matrix [88,93].

Table 4. 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	[33]	Post-industrial	Wood	Fibers	Fibers from local industries
	2017	[36]	Post-industrial	Wood	Flour	Flour of white softwood from local industries
	2018	[39]	Post-industrial	Spruce	Fibers	Fibers from local paper industries
		[40]	Post-industrial	Wood	Sawdust	Furniture manufacturing companies
		[42]	Post-industrial	Pine	Sawdust	Logging in the timber industry
		[43]	Post-industrial	Wood	Sawdust	Woodwork machines from fabrication lab
	2019	[46]	Post-industrial	Eucalyptus, Pine, Hardwood, Maple	Flour	Fibers from timber industries
		[47]	Post-industrial	Spruce	Chips	Lignin extracted from cooking liquor
		[49]	Post-industrial	Pine	Chips	Chips from local sawmill
		[51]	Post-industrial	Teak	Fibers	Waste from local sawmills
		[52]	Post-industrial	Poplar	Fibers	Poplar fibers from milling process
		[54]	Post-industrial	Bamboo	Sawdust	Fibers from local timber industries
		[59]	Post-industrial	Wood	Sawdust	Woodwork machines from fabrication lab
		[61]	Post-industrial	Pine	Fibers	Fibers from local industries
	2020	[67]	Post-industrial	Wood	Flour	Wood flour from furniture industry
		[72]	Post-industrial	Orange wood	Sawdust	Sawdust from orange pruning
		[73]	Post-consumer Post-industrial	Maple, pine	Sawdust	Sawdust from local waste
		[75]	Post-industrial	Oil palm	Fibers	Fruit brunch from local industries
	2021	[79]	Post-industrial	Wood	Flour	Wood flour from furniture industries
		[84]	Post-industrial	Eucalyptus	Sawdust	Sawdust from local industrial waste stream
		[85]	Post-industrial	Wood	Fibers	Fibers from agricultural by-products
Fruit and seed (Plant-derived)	2017	[34]	Post-industrial	Macadamia nut	Shells	Agricultural waste
		[37]	Post-industrial	Cocoa bean	Shells	Raw and unprocessed shell waste
	2019	[57]	Post-industrial	Coffee	Grounds	Oil-extracted spent coffee grounds from supercritical oil extraction
	2020	[65]	Post-industrial	Corn	Cobs	Corn cobs from agricultural waste
		[71]	Post-industrial	Plum seed	Shells	By-products from local waste
		[72]	Post-industrial	Carob, Orange, Tomatoes	Biomass, flour, peels	Scraps from Sicilian cherry tomatoes, discarded carobs, orange peels from pruning
		[73]	Post-consumer Post-industrial	Walnut, cocoa bean, olive	Shells, pomace	Olive pomace, walnut, and cocoa shells from local waste
		[74]	Post-industrial	Soybean	Hull	By-products from local companies

Table 4. Continues.

Waste category	**Year**	**Ref.**	**Waste origin**	**Waste source**	**Waste type**	**Waste supplier**
Fruit and seed (Plant-derived) Continues	2021	[83]	Post-industrial	Cocoa bean	Shells	Cocoa bean shells from local industrial waste
		[86]	Post-consumer	Coffee	Grounds	Spent coffee grounds from local restaurants
		[87]	Post-industrial	Soybean	Hull	Soybean hull from local industries
		[89]	Post-consumer	Coconut	Fibers	Coconut fiber waste from local shops
	2022	[93]	Post-consumer Post-industrial	Banana	Peel	Banana peels from local fruit market
Herbaceous plants (Plant-derived)	2017	[35]	Post-industrial	Rice	Husk	Rice husk from the local industries
	2018	[41]	Post-industrial	Hemp	Shives	Waste from local industries
		[44]	Post-industrial	Rice	Straw	Straw from local farmlands
	2019	[48]	Post-industrial	Sugarcane	Bagasse	Sugarcane bagasse from local mills
		[53]	Post-industrial	Cork	Powder	Residue from cork industry
		[55]	Post-industrial	Hemp	Hurd	Chips from local industries
		[56]	Post-industrial	Flax	Shives	By-products from local cultivations
		[58]	Post-industrial	Sugarcane	Bagasse	Crop sugarcane bagasse from industry
		[63]	Post-industrial	Rice	Husk	Husks from local agricultural industries
	2020	[64]	Post-industrial	Hemp	Stalk	Agricultural biomass from local farmers
		[66]	Post-industrial	Hemp	Shives	Hemp shives from local agricultural waste
		[68]	Post-consumer Post-industrial	Plant	biomass	Cellulose from tissue paper, printing paper, and plant matter
		[69]	Post-industrial	Hemp	Inflorescences, powder	Waste powder from hemp inflorescences
		[70]	Post-industrial	Rice	Husk	Rice husk ash from the local rice mills
		[72]	Post-industrial	Hemp	Leaves	By-products from agricultural waste
	2021	[76]	Post-industrial	Rice	Husk	Rice husk from local rice mills
		[77]	Post-industrial	Rice	Husk	Rice husk from local mills
		[78]	Post-consumer Post-industrial	Opuntia ficus indica, Posidonia oceanica	Cladodes, leaves	Opuntia ficus indica from local industries, Posidonia oceanica from local coasts
		[80]	Post-industrial	Miscanthus	Fibers	By-product from local industry
		[81]	Post-industrial	Buckwheat	Husk	By-product from local industry
		[82]	Post-industrial	Hemp	Biomass	Biomass-fungi mixtures from agricultural biomass of local farmers
		[85]	Post-industrial	Hemp	Fibers	Hemp fibers from agricultural by-products
		[88]	Post-industrial	Sugarcane	Bagasse	Sugarcane bagasse from a local juice shop
		[90]	Post-industrial	Agave	Fibers	Agave fibers from a local tequila company
	2022	[92]	Post-industrial	Kenaf	Stalk	By-product from local industries
		[93]	Post-consumer Post-industrial	Sugarcane	Bagasse	Sugarcane bagasse from local fruit market
		[94]	Post-industrial	Weed grass	Biomass	Weed grass from local agricultural industries
		[95]	Post-industrial	Weed grass	Biomass	Weed grass from local agricultural industries
		[96]	Post-industrial	Opuntia ficus indica	Cladodes	Opuntia ficus indica from local industries
Animal (Animal-derived)	2018	[38]	Post-consumer	Mussels	Shells	Local waste stream
	2019	[50]	Post-industrial	Eels	Skin	Eel skins from the local fish landing center
		[62]	Post-industrial	Croaker	Guts	Farmed Atlantic croaker swim bladder waste
	2020	[73]	Post-consumer Post-industrial	Mussels, eggs	Shells	Mussel shells and eggshells from local waste
Industrial (Plant-derived)	2018	[45]	Post-industrial	Papers	Pulp	Pulp collected from reject press
	2019	[60]	Post-consumer	Paper	Pulp	Office printer paper waste
	2020	[68]	Post-consumer Post-industrial	Paper	Pulp	Cellulose from tissue paper, printing paper, and plant matter
	2021	[91]	Post-consumer	Paper	Pulp	Lab-collected printed office paper

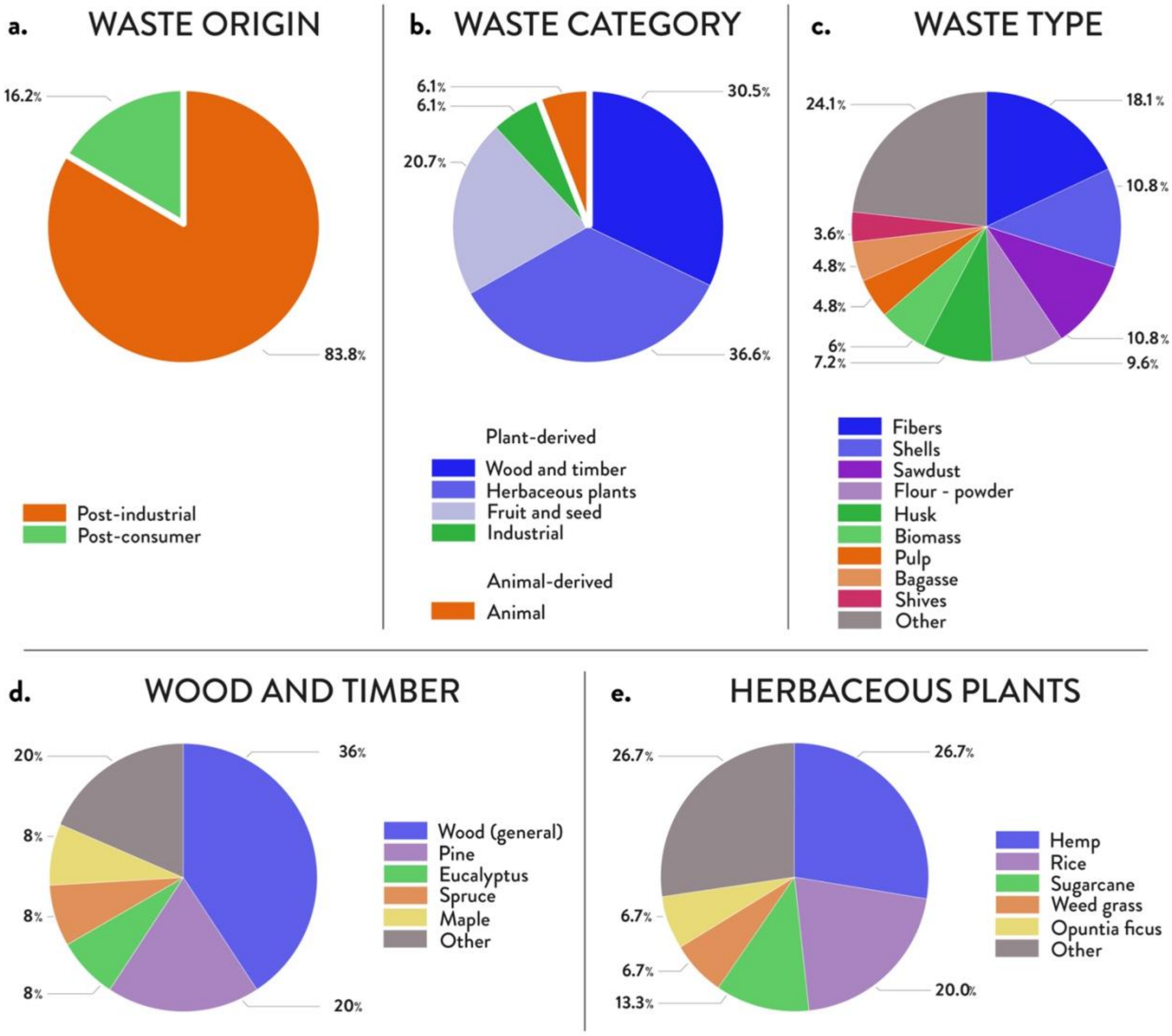

Fig. 3. Analysis of the waste: (a) origin; (b) category; (c) type;
(d) insight on the wood and timber category; (e) and herbaceous plant category.

3.3. New materials

This Sub-section explores the materials used in combination with the biomass waste and/or scraps of the selected works. As shown in Fig. 4a and Table 5, nearly 70% of works extruded thermoplastic polymers together with the 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 [60,78,80,81,90,91,95]. There are also a few examples of polysaccharides used as matrix, especially for cellulose-based waste [43,48,59,68], as well as biomass powder and flour extruded either only together with water [62,65,82,88] or with water and additives, e.g., xyloglucan and guar gum, forming a sort of hydrocolloids [46,93]. Only in three cases, the matrix combined with a waste derived from herbaceous plants was composed of either cement or geopolymer [66,70,92].

Among the thermoplastic materials extruded with biomass waste, PLA is the most popular thermoplastic material used as a matrix in ~54% of selected studies, also considering the blend of PLA with other thermoplastics such as PLA/PBAT and PLA/PHA used to print recycled pinewood fiber and hemp hurd powder reinforced polymers, respectively [55,61]. 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/or scraps were PP (~9%) as well as co-polyesters (CoPE, ~9%), and ABS (~7%). The choice of these thermoplastic materials was probably 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 (nearly 70% of the papers) are in the range of 1-29% wt. [33,39,54,72]. In a few studies (~3%), a waste percentage of 40-44% was also added and tested, for example in the case of extrusion of wood waste with PLA and with recycled HDPE [40,42].

However, a higher amount can induce a detrimental effect on the mechanical properties of the wood fiber reinforced thermoplastic [52]. 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 [38,73], sugarcane bagasse powder without the use of any other additive [88] and 3D printed hemicellulose waste [65].

Table 5. 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	[33]	PLA/PHA blend	Wood fibers (shredded)	30
	2017	[34]	ABS	Macadamia nutshell fibers (shredded)	19-29
		[35]	PBAT	Silica/carbon hybrid nanoparticles	0.5-1.5
		[37]	PCL	Cocoa shell powder	10-50
	2018	[39]	BioPE	Thermomechanical pulp fibers	10-20
		[40]	PLA	Wood waste	30
		[41]	PLA	Hemp powders	1-5
		[42]	Recycled HDPE	Wood sawdust	15
		[44]	ABS	Rice straw flour	5-20
		[45]	BioPE	Thermomechanical pulp	10-30
	2019	[47]	PLA	Lignin	20-40
		[49]	PHB	Biorefinery lignin	10-50
		[51]	PLA	Teak wood fibers	1-5
		[52]	PLA	Poplar fibers	20
		[53]	TPU	Cork powder	1-5
		[54]	PLA	Bamboo fibers	10-15
		[55]	PLA/PBAT blend	Hemp hurd powder	8-27
		[56]	PLLA, PBAT, PLLA/PBS blend	Flax fibers	10-30
		[57]	PLA	Spent coffee grounds	5-20
		[58]	PLA	Sugarcane bagasse fibers	3-15
		[60]	Recycled PP	Cellulose	5-20
		[61]	PLA/PHA blend	Pinewood fibers	30
		[63]	PLA	Rice husk or wood flour	10
	2020	[67]	PLA	Wood flour	1-7
		[69]	PLA	Hemp inflorescences	10-25
		[71]	PLA/PHB blend	Cellulose nanocrystals	1
		[72]	PLA	Hemp biomass, cherry tomatoes scraps, carob flour, or orange pruning	10-25
		[74]	Co-PE	Soybean hull	5-10
		[75]	ABS	Oil palm fibers	5
	2021	[77]	Recycled PP	Rice husk (Shredded)	5-10
		[78]	PLA	Opuntia ficus indica or Posidonia oceanica	10-20
		[79]	PLA	Wood flour	5
		[80]	PTT	Miscanthus biocarbon	5-10
		[81]	PLA/PBAT blend	Buckwheat husk	5-10
		[83]	Recycled PP	Cocoa bean shell (Shredded)	5
		[84]	PLA/PBS blend	Cellulose nanofibers	1-5
		[85]	PLA	Hemp fibers, wood fibers	20-30
		[87]	CoPE	Soybean hull fibers	5-10
		[89]	PP	Coconut fibers	10-50

Table 5. Continues.

Material	**Year**	**Ref.**	**Matrix**	**Filler/reinforcement from waste/by-products**	**% wt.**
Thermoplastic composite (continues)		[90]	PLA	Agave fibers (Shredded)	3-10
		[91]	PLA	Paper pulp	5-15
	2022	[94]	CoPE	Weed grass (Shredded)	5-20
		[95]	PLA	Weed grass (Shredded)	10-20
		[96]	CoPE	Opuntia ficus indica (Shredded)	10
Thermoset composite	2017	[36]	Urea formaldehyde	Wood flour	13
	2021	[86]	Epoxy resin	Biochar	1-3
Ceramic composite	2020	[66]	Concrete	Hemp shives	30
		[70]	Concrete	Rice husk ash	20
	2022	[92]	Geopolymer	Shredded kenaf fibers	3
Polysaccharide composite	2018	[43]	Chitin	Cellulose (fungus-like material)	12
	2019	[48]	Alginate	Cellulose nanofibrils	20
		[50]	Alginate	Eel skin collagen	9-23
		[59]	Chitin	Cellulose (fungus-like material from [43])	9-50
	2020	[68]	Chitin	Cellulose	n.a.
		[73]	Sodium alginate	Mussel shell or eggshell powder, walnut shell or cocoa shell powder, olive pomace, pine, or maple sawdust	15-61
Hydrocolloids	2018	[38]	Sugar, water	Mussel shell powder	70
	2019	[46]	Water, Xyloglucan	Cellulose nanocrystals, wood fibers	5-30
	2021	[76]	Water, guar gum	Rice husk	42
	2022	[93]	Water, guar gum	Sugarcane bagasse and banana peels flour	32
Water-based pastes	2019	[62]	Water	Swim bladder powder	4.8
	2020	[64]	water	Biomass, psyllium husk powder (Mycelium)	19
		[65]	water	Hemi cellulosic paste	86
	2021	[82]	Water	Biomass, wheat flour, psyllium husk powder	18
		[88]	Water	Sugarcane bagasse powder	53

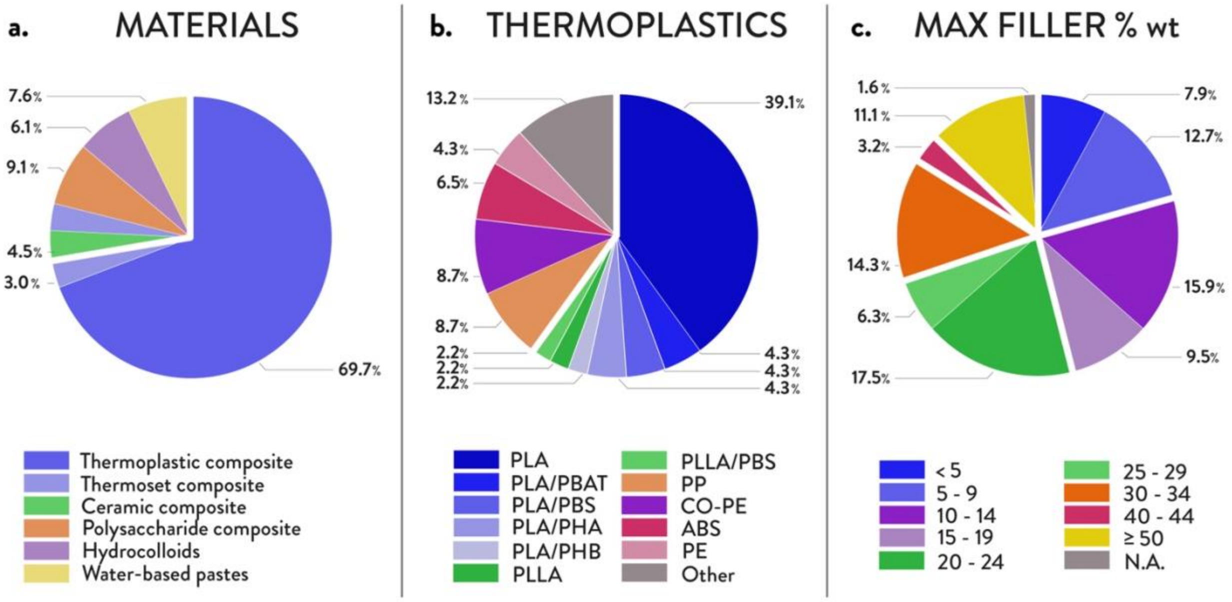

Fig. 4. Analysis of the new 3D printable materials: (a) composite category; (b) thermoplastic polymer-based matrix materials; (c) weight percentages of filler/reinforcement from the waste source.

3.4. Extrusion-based AM processes

This Sub-section focuses on the analysis of 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 [19], 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 to 3D print 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. [38,53,65].

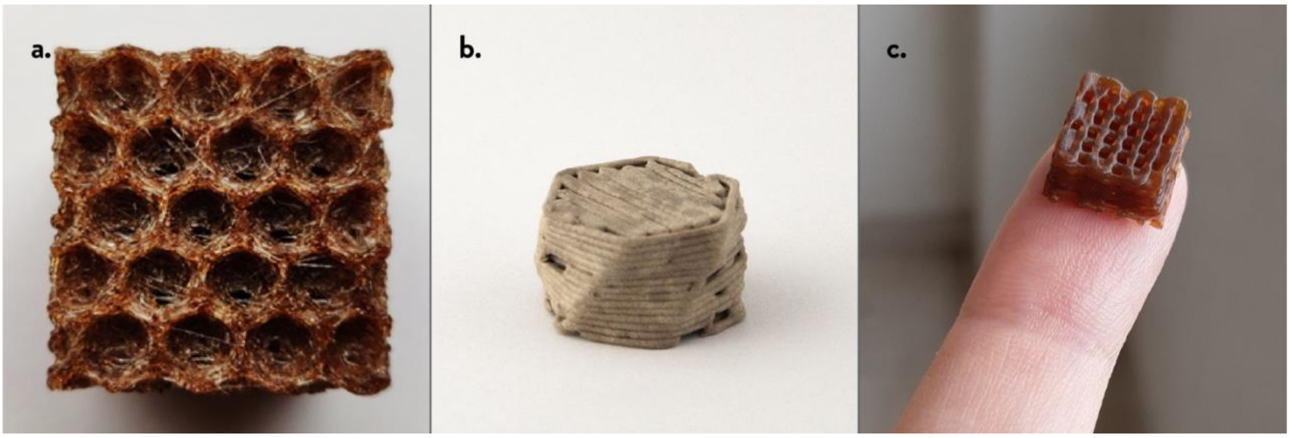

Fig. 5. 3D printed materials from biomass waste: (a) cork/TPU composite foams [53] (FFF, Reprinted with the permission of Elsevier); (b) mussel shells/sugar water composite [38] (DIW, Reprinted with the permission of Elsevier); (c) hemicellulosic paste from corn cobs [65] (DIW).

According to Table 6 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 kinds of 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 [17]. However, DIW can process a wider range of material categories than FFF and, consequently, it is a valid option for all those materials, and biomasses, which are not 3D printable by using FFF 3D printers, i.e., hydrocolloids or concrete filled with biomass [70].

As shown in Fig. 6a, more than 85% 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 be not true for DIW processes. Processing different material categories also means to tackle with various potential applications and dealing with different orders of magnitude. The works focused on DIW processes show a bigger variation in the 3D printer format with respect to FFF-related publications. Almost one-third of the DIW 3D printers are large format systems, whereas only 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 wider range of applications, from small format bioprinters to large format structures for the building sector [48,50,59,68]. Moreover, small format FFF 3D printers often use filaments as primary feedstock, adding further reprocessing to obtain new 3D printable materials [28]. These steps may increase the reprocessing time or cause degradation of the raw materials, i.e., through further heating cycles for filament extrusion.

Similarly, the development of 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 itself [76,88,93,98] or derived from previous works of different research groups [66,99]. From this review, DIW custom 3D printers are seven times as many FFF custom systems, and they represent 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 [100,101]. Hence, custom systems have been developed to improve the control of the 3D printing parameters of these new materials for DIW, defining ad-hoc solutions to process viscous pastes at room temperature, i.e., by using screw extruders or pump systems on robot arms [43,73,92]. 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 [17,20,24,28].

These differences are also visible from the main 3D printing parameters reported in Table 6. Using DIW processes often means dealing with bigger nozzle diameters, lower speeds, and thicker layers compared to FFF. On the one hand, these specifics result in lower 3D printing resolutions but, on the other hand, in a wider range of 3D printable materials and recovered biomass waste and/or by-products. 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 6. 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	[33]	//	//	18	//	Prusa i3 Rework
	2017	Small	[34]	//	0.5	//	//	Leapfrog Creatr
		Small	[37]	//	0.6	50	0.3	Prusa i3 Hephestos
	2018	Small	[39]	//	0.4	15	//	Ultimaker Original
			[41]	//	//	//	0.1	Ultimaker 3
			[44]	//	1	//	0.2	Printrbot Simple Metal
			[45]	//	0.4	15	//	Original Prusa i3
		Large and small	[40]	//	0.5	62.5	0.15	Delta-type RepRap, Re:3D Gigabot version GB2
			[42]	Robot arm, custom	//	30	0.3	High-speed single screw extruder on a robot arm, and Delta 3D printer
	2019	Small	[47]	//	0.4	35	//	Prusa i3 MK3S
			[49]	//	0.75	25	0.3	MakerGear V2
			[51]	//	0.4	//	0.2	MakerBot Replicator Z18
			[53]	//	//	60	0.2	Anycubic Chiron
			[54]	//	//	20	//	low-end 3D printer after nozzle adaptation
			[55]	//	0.4	60	0.15	Da Vinci 1.0 Professional
			[56]	//	1	13-25	//	Prusa i3 Rework
			[57]	//	0.4	//	//	Einstart-S
			[58]	//	0.6	40	0.1	S1Architect
			[60]	//	0.8	20-50	0.2	Lulzbot Taz 6 MEAM
			[61]	//	0.4	23	0.2	Prusa i3
			[62]	//	0.4	30	0.2	Shenzhen 605 S model
			[63]	//	0.75	//	//	Makergear M2
		Large	[52]	//	7.62	//	5.84	Big Area Additive Manufacturing (BAAM) system at Oak Ridge National Laboratory
	2020	Small	[67]	//	0.4	//	0.1	Flythinking FS-14
			[69]	//	0.6	60	0.25	D300 Technology®
			[71]	//	0.4	33	//	WASP Delta 2040 Turbo 2
			[72]	//	0.6	//	//	D300 Technology®
			[74]	//	0.5	30	0.2	Desktop Printrbot
			[75]	//	//	//	//	UP plus 2 model
	2021	Small	[77]	//	0.8	60	0.25	Fusedformcorp 3D FF STD Doppia
			[78]	//	//	45	0.1	Sharebot Next Generation
			[79]	//	0.4	50	0.4	FS-200 printer (Guangzhou Flythinking Co)
			[80]	//	0.5	35	0.3	Lulzbot Taz 6
			[81]	//	0.8	50-80	0.15	Prusa i3 MK3
			[83]	//	0.8	60	0.25	Fusedformcorp 3D FF STD Doppia machine
			[84]	//	0.4	60	0.3	Wanhao Duplicator i3 plus
			[85]	//	//	//	0.1	Sharebot NG 2
			[87]	//	//	//	//	Prusa i3 systems

Table 6. Continues.

Extr-based AM	Year	Printer format	Ref.	Other specs	Nozzle (mm)	Speed (mm/s)	Layer h. (mm)	3D Printer
FDM – FFF (Continues.)	2021 (C.)	Small (C.)	[89]	//	//	40	0.5	Anycubic Chiron
			[90]	//	//	50	0.3	Wanhao Duplicator 4
			[91]	//	0.4	30	0.2	Shenzhen 603S model
	2022	Small	[94]	//	//	50	0.1	Next Generation - Sharebot
			[95]	//	//	20	0.1-0.2	Next Generation - Sharebot
			[96]	//	0.4	50	0.1	Sharebot Next Generation
LDM - DIW	2017	Small	[35]	Custom	//	//	//	Hyrel system 30 M model
			[36]	//	//	15	//	Fisnar robot 7400; a 3-axis robot
	2018	Small	[38]	Custom	0.84	5	1	Modified Ultimaker 2+
		Large	[43]	Robot arm, custom	7	//	3	Six-axis articulated industrial robot with a precision material dispenser, and a material pump system
	2019	Small	[46]	Custom	1.291	5	//	Modified Hyrel3D 30M with a syringe extruder
			[48]	Bioprinting	0.58	3	//	Regemat3D printing unit (version 1.0)
			[50]	Bioprinting	0.5	//	//	BIOBOT Allevi
		Large	[59]	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	[64]	//	4	//	//	Delta wasp 2040 clay mode
			[65]	Custom	0.54-0.68	//	//	Custom-made 3D printer built with 3D4E
			[73]	Custom	1.6	6	1.1	Ultimaker 2+ modified for paste printing with the Stoneflower Ceramic matrix 3D Printing KIT Basic
		Large	[66]	Custom	20	//	5-10	Custom-made 3D printer extruder inspired by [99]
			[70]	//	//	12.5	//	Custom screw-type extruder mounted on a three-axis gantry system, Kenyo 3D printer
		Large and small	[68]	Robot arm, custom	1-6.5	50-150	//	Custom-made apparatus (small and large format)
	2021	Small	[76]	Food printing, custom	0.82	//	//	Custom delta (Controlled Additive-manufacturing Robotic Kit) by [98]
			[82]	//	6	30	6	Delta WASP 2040 clay
			[86]	//	0.48	50	0.4	Hyrel-30M benchtop
			[88]	Food printing, custom	0.82	//	//	Custom delta (Controlled Additive-manufacturing Robotic Kit) by [98]
	2022	Small	[93]	Food printing, custom	1.2	7-12	//	Custom delta (Controlled Additive-manufacturing Robotic Kit) by [98]
		Large	[92]	Custom	11-28	//	//	Self-made apparatus with different nozzle sections

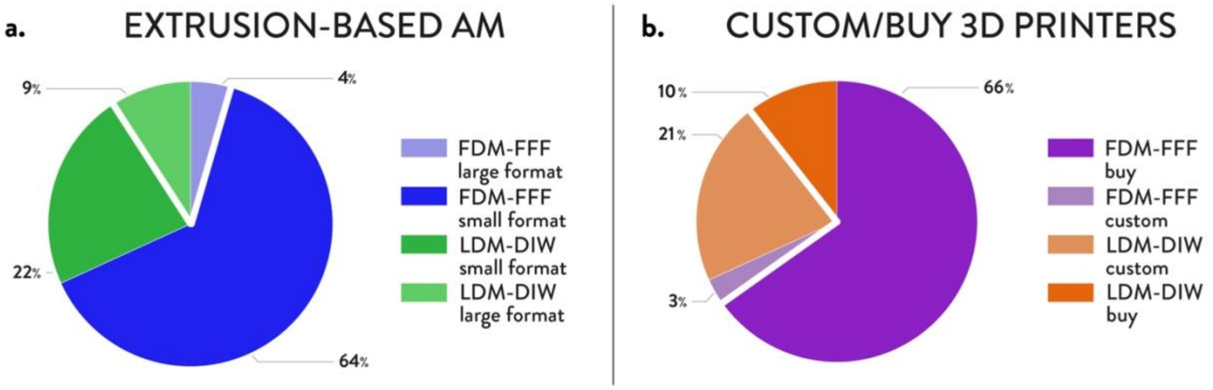

Fig. 6. Analysis of the extrusion-based AM processes: (a) process and printer format; (b) custom/buy 3d printer systems.

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/or 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 are mainly focused 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 better highlight some possible trends for the use of 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 includes some hypothesis on possible fields of applications or potential uses (~53%), 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 the 3D printable materials within the real context. The list of the specific products and/or applications and the general fields is visible in Table 7. As from the previous Sub-section, more applications are linked with the FFF process (13 works), confirming its prevalent use with respect to DIW systems (9 works). Despite the limited number of applications, six main fields were defined to cluster the publications with a specific application or demonstration ("buildings and architecture", "furniture", "bioengineering", "equipment", "prosthetics and medical", and "packaging"). As shown in Fig. 7b, the most represented fields of application are "buildings and architecture" and "furniture", representing nearly one-fourth and one-fifth 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 [40,73], to large architectural structures, i.e., sculptures (Fig. 8a) and turbine blades [59,68], 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) [50] or "prosthetics and medical" focused on customized orthoses (FFF) [72].

Table 7. 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	[40]	Furniture	Desk cable feedthrough parts, drawer knob
	LDM - DIW	[38]	Equipment	Soil fertilizer flowerpot
		[43]	Buildings, architecture	Wind blade turbine
2019	FDM - FFF	[47]	Other	Smartphone protective case
		[52]	Furniture	Podium base
		[54]	Equipment	Brake levers of a bike
		[61]	Equipment	Technical part for 3D printing
		[62]	Other	Moisture actuators for meteor sensitive architectures, biomedical devices, soft robotics
	LDM - DIW	[46]	Buildings, architecture	Mashrabiya, a wooden window (Patent application)
		[50]	Bioengineering	Scaffold for tissue fabrication
		[59]	Buildings, architecture	Wind turbine blade
2020	FDM - FFF	[67]	Furniture	Scaled chair model
		[68]	Buildings, architecture	Sculpture (Hydra)
		[69]	Prosthetics, medical	Neck orthosis
		[72]	Prosthetics, medical	Customized neck orthosis and laryngoscope
	LDM - DIW	[65]	Bioengineering	Scaffold prototype for biomedical application
		[66]	Bioengineering	Scaffold prototype for biomedical application
		[73]	Furniture, accessories	Lamp, Hairpin
2021	FDM - FFF	[84]	Packaging	Main body of a fruit crate and compostable packaging
		[85]	Buildings, architecture	Geogrids for Construction and Demolition sector
		[96]	Other	Controlled release fertilizers for agriculture
	LDM - DIW	[88]	Packaging	Customized food package casings

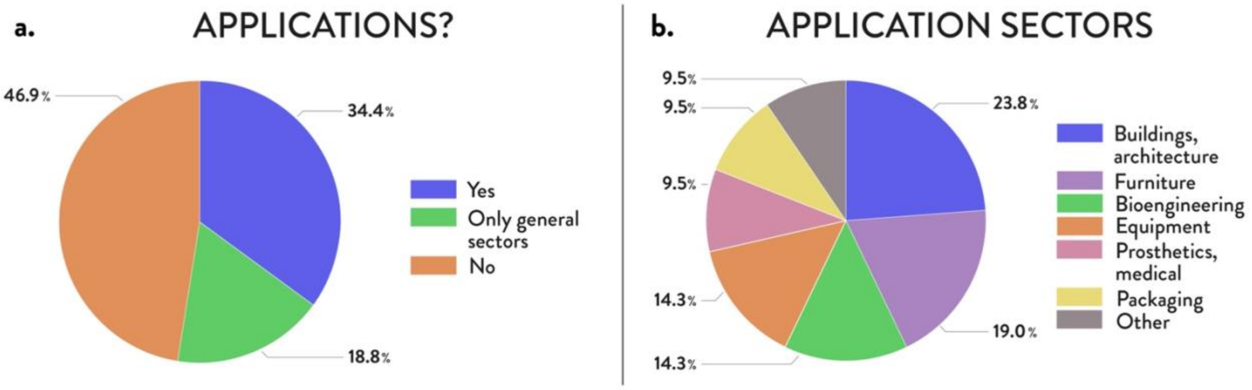

Fig. 7. Analysis of the applications from the selected works: (a) development of applications; (b) main application sectors.

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/or by-products as the main source for new 3D printable materials. Three main trends may be detected: production and selling of commercial materials; production and selling 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 [102]. Other industrial realities allow not only to buy the 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) [103]. In addition, emerging design studios and professionals are developing new products starting from biomass materials following the approach of Material Driven Design (MDD) [104]. The design studio "Krill Design" has been developing new filaments for FFF in collaboration with agri-food industries starting from their biomass waste and/or by-products, i.e., orange and lemon peels, using these materials for new products such as "Ohmie – the Orange Lamp" (Fig. 8c) [105].

These exemplary best practices show possible synergies between different industry stakeholders to create new circular economy strategies from the upcycling of biomass waste, fostering the real exploitation of these materials. Some connections between academic research and practical contexts may be possible by fostering the knowledge of these new materials amongst the design practitioners and industrial realities for their use in real applications [17].

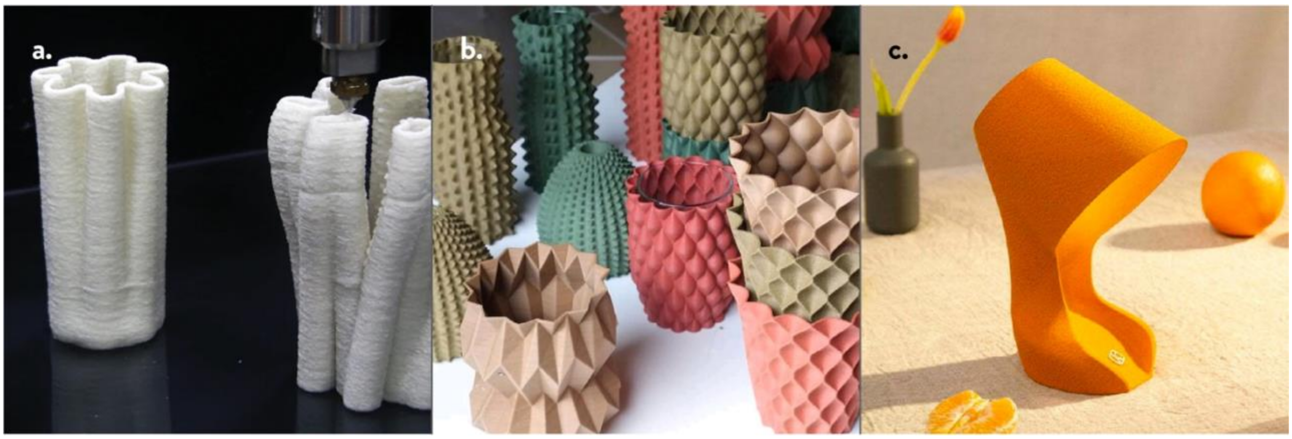

Fig. 8. Examples of 3D printed applications from biomass waste: (a) "Natural Composite Pillar" made of chitin/cellulose composite through large format DIW [68]; (b) "Hemprinted" e-shop of products and filaments for FFF made of PLA filled with hemp, tomato, and pomegranate waste [103]; (c) "Ohmie Lamp" made of PHB filled with orange peels through FFF by Krill Design [105].

4. Conclusions

This review systematically collected the works dealing with the development of 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 circular economy focusing on the raw materials, the new 3D printable materials, the 3D printing specifics, and the possible applications. 64 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 (50 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/or 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/or by-products recycled 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 the presence of defects in the final 3D printed objects. For this reason, mechanical grinding of biomass waste was carried out in numerous works before the preparation of 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 biodegradability. Other thermoplastics such as PP, ABS, and copolyesters were also used to a certain extent, probably due to their popularity as 3D printable materials, low cost, and good mechanical properties. There are also several works, which combined bio-waste with polysaccharides, like alginate and chitin, as well as with water and other additives, likely 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 produce a negative effect on 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 for its capability 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 the exploitations of 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/or by-products through AM, ranging from the production of commercial filaments to the design of new kinds of products. However, future research should strengthen their real exploitation, i.e., fostering the knowledge of these emerging materials amongst the design professionals and the interested stakeholders. 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.

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.

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