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Review

Additive manufacturing of natural materials

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ABSTRACT

As additive manufacturing (AM) technology continues to advance for computer-aided design and engineering applications, a parallel imperative emerges — a conscientious shift towards more responsible material practices, aligning with ethical, environmental, and social sustainability considerations. The present systematic review analyzes the state-of-the-art developments in relation to AM using natural, low-carbon, and readily available material practices. The results show that published work is situated at the intersection of material science, digital fabrication, and construction, with an array of geo-, bio-, and living mix designs, and different properties analyzed. Under certain conditions, a move towards more use of natural materials could be the solution to source more responsible materials while contributing to the quality of the built environment and the planet Earth itself. The long-term contribution is to provide leading guidance for future research aimed at developing novel and bespoke natural materials in digital fabrication and advanced manufacturing.

1. Introduction

Computational design must deeply engage in sustainable material development to foster profound environmental and societal transformations in digital fabrication. The clock is ticking—irreversible climate change is approaching if change does not happen. Human- and machine-made built environments have been altering natural landscapes, and these alterations have largely triggered and exacerbated biodiversity and climate emergencies, disproportionately impacting underrepresented communities [102], particularly in regions where natural materials are a traditional trade. In parallel, building operations and construction account for a significant percentage of global greenhouse gas emissions, contributing to an estimated 37% of global energy- and process-related carbon dioxide (CO_2) emissions in 2021 [110]. Additionally, over 10 billion tonnes of construction and demolition waste is produced annually globally, which occupies 35–65% of global landfills [68].

Additive manufacturing (AM), also referred to as rapid prototyping or 3D printing, exhibits a multiplicity of advantages. AM can reduce the amount of material needed and, as a consequence, the waste generation typical to subtractive manufacturing methods [11]. AM represents one of the most promising innovations with digital and generative design possibilities and the potential to enhance global supply chain capabilities while reducing high-risk, labor-intensive jobs [10,87]. The advent of AM technologies through digital fabrication has primarily been motivated by the demand for increased productivity and customization while reducing material waste in all sectors, including manufacturing and architecture ([16]; García [53]).

To date, a growing number of entire buildings are constructed using AM. Notable examples include the 3D Printed Urban Cabin 3D by DUS Architects [41], the DFAB House by Gramazio, and Kohler Research [88]. Segments or portions of structures are commercially 3D printed mostly utilizing cementitious mortar as the main material [30,62,64]. However, the impact of 3D-printed concrete is almost twice as large as that of the same quantity of casted concrete, given the following two main reasons: firstly, the significantly greater higher content of cement within 3D printed concrete compared to conventional concrete, and secondly, the higher energy intensity of the robotic deposition processes [9].

To combine advanced computing and environmental sustainability, one viable pathway to addressing these intricate environmental and societal challenges by the agency of AM is using climate-specific and low-carbon natural building materials that are locally available, minimally processed, healthy, and community-engaging [14]. Indeed, recent studies demonstrate the efficiency of substituting climate-intensive materials with bio- and earth-based building ones that are environmentally and socially healthier than conventional highly processed cement- and petroleum-based materials [21,28,42,92,94]. Specifically for natural materials, a few examples of large-scale earth structures have been 3D printed starting in 2015. Pylos projects represent the first

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precedent [63], followed by the Gaia [25] and Tecla [5] houses by WASP, an Italian manufacturer of 3D printers, and Mud Frontiers [44], which employed large printers directly on site.

Natural materials are defined as substances and composites that are minimally processed and nontoxic by using readily available sources on or around the construction site. Due to their nontoxicity, traditional nature, and adaptability to various shapes, natural building materials are often reminiscences of Indigenous wisdom for architectural techniques [101]. They include a range of naturally occurring constituencies, but can be summarized into two base constituencies comprising *geological* products such as earth, sand, larger aggregate, and stone; and *biological* products like mycelium, vegetation plants, plant-like protists for instance algae, agro-waste, and vegetable fiber by-products [13]. This range of substances can be combined into mix designs using construction methods from traditional techniques such as adobe, rammed earth, compressed earth blocks, cob, earthbags, straw-bale construction, and light straw clay, but that can be implemented in more advanced construction techniques.

Advanced fabrication technologies can –and should – help in fighting resource scarcity by enabling material efficiency and low-volume manufacturing during the fabrication process while assuring, or even ameliorating, their technical performances [78]. To further adopt this concept, there is a need to integrate the fields of materials, hardware, and software development, through a systematic review. With these topics, this work asks the following research questions: what are the materials used and the steps to create mixtures adequate for digital fabrication? What are the possible applications and performances of existing demonstrations? This systematic review aims to identify and systematically categorize the state-of-the-art of how natural materials - new versions of ancient practices - have been reinterpreted with AM and provide the essential tool to further their adoption in contemporary architecture.

2. Research objective, scope, and methodology

The objective of this systematic review is to critically map the terrain of additive manufacturing of natural materials in terms of material mixtures, techniques, additives, and performance of the final printed element. The scope includes applications within the construction and architectural manufacturing fields with earth- and bio-based material mix designs. The review methodology, outlined in Fig. 1, was developed to ensure a systematic search process and comprehensive review of the relevant literature [109], by following the PRISMA protocol [85], which includes the Identification, Screening, Eligibility and Inclusion phases. First, the framework and search strategy were identified. Then, relevant literature was evaluated according to three screening phases: i) database search, ii) title and abstract screening, and iii) full paper screening and categorization within a database that resides in an Excel spreadsheet. Lastly, the eligible data were analyzed and discussed according to outlined criteria while iteratively identifying additional papers cited within the collected ones. The systematic search has been performed in established database and indexes including ScienceDirect, Taylor & Francis, as they contained a wide selection of scientific articles, and IOP Science and CumInCAD, which collect the proceedings of recognized conferences in the filed of additive manufacturing and digital fabrication for architecture, by using the following keyword combinations: "3D printing" AND "natural materials" AND "construction materials", "3D printing" AND "earth materials" AND "construction materials", "3D

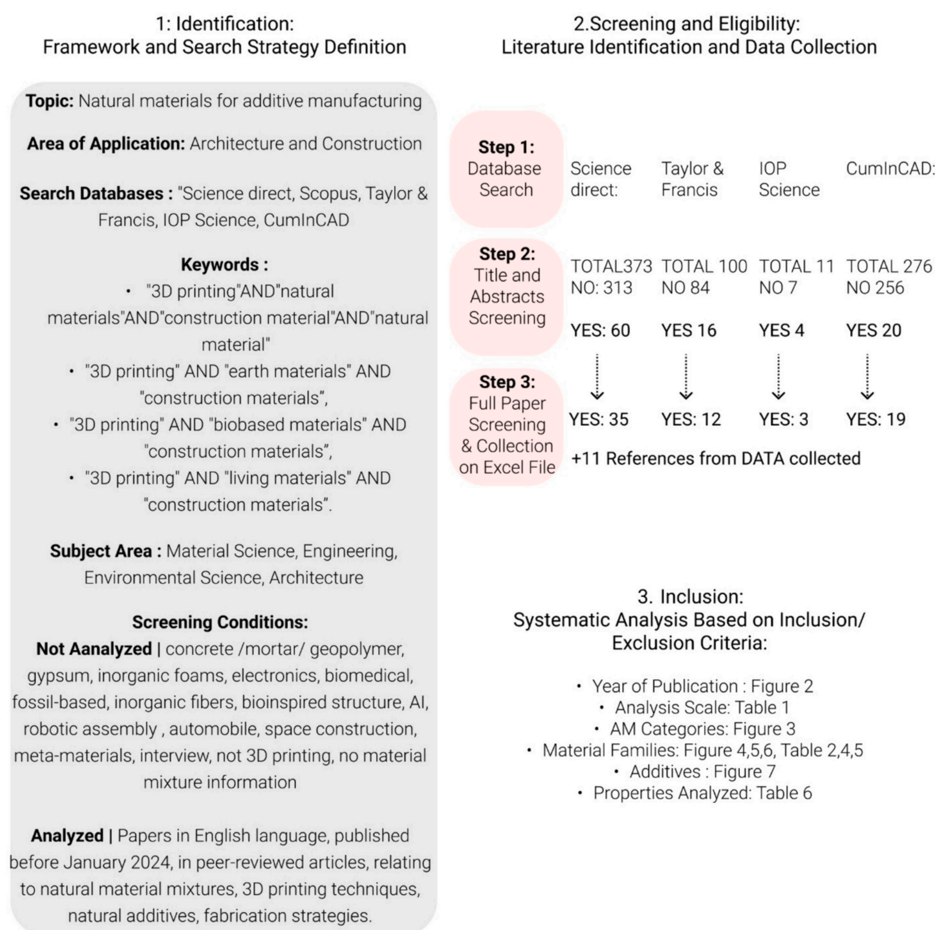

Fig. 1. Systematic review methodology incorporated in this paper.

printing" AND "biobased materials" AND "construction materials", and "3D printing" AND "living materials" AND "construction materials".

3. Results

3.1. General observations

The preliminary literature search has resulted in 614 academic papers identified between 1995 and April 2024 that focused on the use of natural materials in the fields of architecture and construction. This time frame was selected to grasp the most contemporary research practices on this emerging topic. Following the three-step screening process, 80 papers were deemed relevant and were fully reviewed and documented in an Excel database (Appendix A), serving as the resource for the systematic analysis herein. The data obtained from the papers were categorized into five main groups focusing on 1) material families (e.g., earth-based, clay-based, and fiber-based, etc.); 2) machines and AM technologies utilized (e.g., material extrusion, binder jetting, etc.); 3) mix-designs; and 4) performance analyzed (e.g., structural, geometrical, environmental, etc.). The focus was only on peer-reviewed journal articles, book chapters, and conference contributions.

The identified papers were found to belong to the domains of architecture, material science, engineering, and environmental science. Fig. 2 illustrates the number of papers identified according to the year of publication, which may indicate the emerging publication trend and growing interest in the topic, especially since 2018.

Research in this field grew slowly between 1995 and 2014. Prior to 2014, only one publication focused on biomedical engineering was found, indicating that the interest in these topics emerged in the paper repositories from 2014 onward. The publication trend can be divided into three main time periods. The first period, from 2014 to 2015, corresponds to the time of Neri Oxman [40]; [89] and their pioneer research focus at the intersection of biology and digital fabrication. In the same year, it was also released a research on robotic deposition of clay [52]. As evidenced from 2017 onwards, some of the research interest in digital design shifted towards a more multidisciplinary approach and on material extrusions at room temperature to work with clay/soil pastes and natural compounds that do not required heat to be extruded from nozzles. During the second period, from 2017 to 2022, various conferences such as ICSA 2022 included "Ecology of structures and architecture" in their themes, leading to a significant increase in publications on the subject. The number of publications on the topic gradually increased, peaking in 2022, with papers focusing on various natural materials.

Table 1 details the analyzed publications according to their research focus, or building scale analysis, enumerating the number of publications per each journal or conference found, providing guiding insights into predominant publication venues for future researchers in the field. The largest research scale identified is shown to be confined to a building component or pavilion scale. Looking into the established examples found in the literature, the review identifies that real- and large-scale 3D printed buildings with natural materials, such as the first 3D printed earth-based houses, Gaia and Tecla house [25], were realized as applied demonstrations by educational and industry leaders such as WASP and Institute for Advanced Architecture of Catalonia (IACC). With exception of the Tecla house [48], these examples were not elaborated within scientific publications, and therefore were not established as a peer-reviewed academic research. The variety of repository types (Journals, Conferences, and Books) summarized in Table 1 suggests the multidisciplinary of this field, which is tackled from a range of perspectives. Papers with authors from material studies are shown to identify mix-design ingredients and possible additives or fabrication strategies to obtain extrudable and buildable 3D-printed outcomes. Papers authored by authors from biomedical fields were not included in

Table 1
Publication analysis according to research focus and building scale. In bold are the most prominent journals by publication items.

Research focus	Number of Papers Identified According to Journal / *Book* / Conference	Reference
Building Materials (29)	Powder Technology (1), Journal of the European Ceramic Society (1), **Construction and Building Materials (7),** Materials & Design (1), International Conference on Structures and Architecture ICSA (6), eCAADe (4), Architectural Science Review (1), Biomimetics (1), Manufacturing Letters (1), CAADRIA (2), ICBBM (1), Materials (1), NOCMAT (1), Journal of Building Engineering (1), *Handbook of Natural Polymers, Volume 1 (1)*	[106], [79], [4], [105], [49], [12], [7], [20], [112], [71], [100], [84], [17], [19], [1], [23], [75], [111], [2], [67], [104], [48], [91], [18], [8], [38], [26], [22], [115]
Building Components (31)	Construction and Building Materials (3), Materials Today Communications (1), Ecological Engineering (1), Journal of Building Engineering (1), Materials Today: Proceedings (1), Architectural Science Review (2), Towards a Robotic Architecture (1), Computer-Aided Design (1), **eCAADe (5), ACADIA (6),** CAADRIA (3), Cement and Concrete Research (1), Materials & Design (1), Fabricate (2), Automation in Construction (1), Journal of Cleaner Production (1)	[46], [98], [15], [43], [37], [56], [32], [39], [40], [24], [29], [31], [35], [47], [52], [60], [66], [99], [69], [70], [72], [83], [103], [50], [54], [55], [89], [58,59], [3], [58,59], [33]
Review Papers (19)	Birkhäuser (1), Journal of Cleaner Production (1), Materials Science and Engineering: R: Reports (2), Processing Technology for Bio-Based Polymers (1), Progress in Materials Science (1), Heliyon (1), Rethinking building skins (1), Renewable and Sustainable Energy Review (1), Advanced Manufacturing Processes (1), Bioinspiration & Biomimetics (2), Biomimetics (1), Journal of Physics: Materials (1), Automation in Construction (1), Frontiers of Architectural Research (1), **Construction and Building Materials (2)**,Matter (1)	[116], [57], [117–121], [87], [122–128], [108], [129], [6], [130]
Architectural Theory (1)	Planning Theory & Practice (1)	[131]

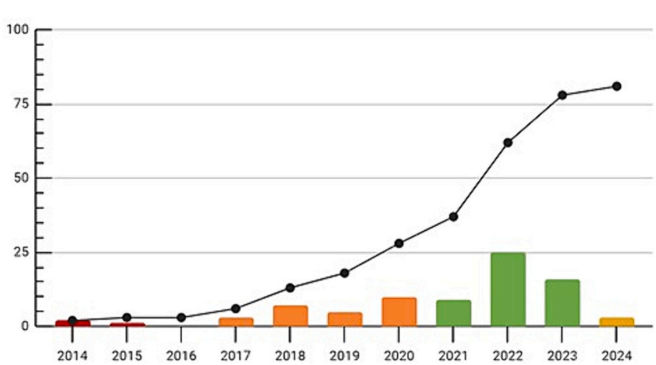

Fig. 2. Number of publications identified according to the publication year.

this review; however, interestingly enough, these papers were found to be mainly focused on the creation of bioinspired patterns and design inspirations to optimize the material use and strategically deposit it where its performance requires [74,93,113]. Lastly, papers with authors from architecture and civil engineering fields are mostly focused on the properties and design freedom of printed building elements, comparing them with more traditional technologies and materials. The specific themes of the review papers inserted in Table 1 can be found in Appendix A under "Analysis type" column.

3.2. Additive manufacturing techniques for natural materials

According to the American Society for Testing and Materials (ASTM) standards, AM technology "applies the additive shaping principle and thereby builds physical three-dimensional (3D) geometries by successive addition of material" [65] using one or more of the following seven main process categories: (1) Binder jetting (BJT), (2) Material jetting (MJT); (3) Material extrusion (ME); (4) Direct energy deposition (DED); (5) Sheet lamination (SHL); (6) Vat photopolymerization (VPP); (7) Powder bed.

Illustrated in Fig. 3, of these seven categories, the identified papers were shown to predominantly focus on material extrusion (57 papers). A few (4 papers) were shown to focus on binder jetting. Only one paper was shown to focus on power bed fusion techniques and was thus taken out of the scope of this analysis.

Material extrusion (ME) involves the deposition of a material from an extruder and it is usually referred to as a "heating mechanism that melts a thermoplastic filament" [90]. Nevertheless, ME processes can also be applied to viscous materials like clay- and earth-based materials without the need for thermal heat at the moment of deposition [86]. Some authors further define techniques utilized within the ME framework to specify the peculiarity of extrusion involved without heat, specifically, extrusion-based ME processes with highly concentrated suspensions made with powders [73]. This may include robocasting/direct ink writing (DIW) [43], paste extrusion modeling (PEM), and other paste-based technologies [19,20,49,112]. Similarly, flow-based fabrication

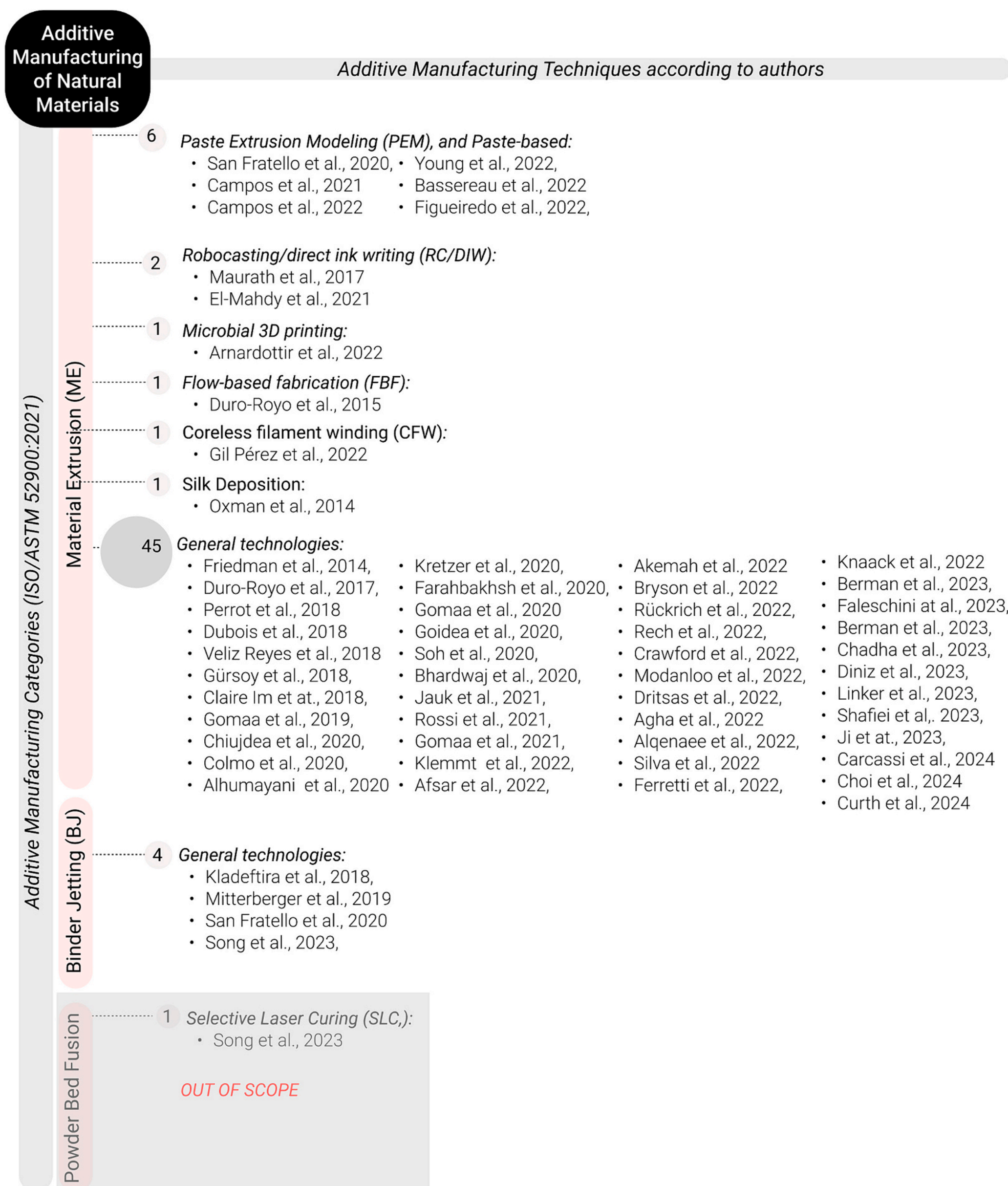

Fig. 3. Identified literature on natural materials in additive manufacturing, categorized according to the ASTM Additive Manufacturing categories (Material Extrusion, Binder Jetting, and sub-category technologies observed in the literature).

(FBF) enables the production of functionally graded materials through the flow control of syringes containing different water-based substances. Other processes make use of living organisms such as microbial printing [7] and silk deposition [89]. Lastly, coreless filament winding [54] was classified by the authors.

The printing process of Binder Jetting, which originated at MIT in the early 1990s [45], involves the application of a liquid binder onto a thin layer of powder through spraying or jetting [27]. The binder solidifies the powder, and subsequent layers of powder are successively added, repeating the process numerous times. It can be used with gypsum, sand, glass, metal, and similar materials [86]. [106], used both binder jetting (BJ) and selective laser curing (SLC) technologies with silica sand materials [106]. SLC technology is based on a selective laser sintering (SLS) process where a powder material is heated to near-melting temperatures (sintering), to bond together particles and obtain a solid element [107]. Although significantly lower temperatures are applied in SLC (less than 400 °C), it was still taken out of the scope of this review, given the desired focus on readily available materials and accessible technologies with minimum energy demand [51].

3.3. Mix designs of natural materials for printability

Corresponding to the broader definition of natural materials articulated in the Introduction section, three main material families were identified as currently applied in AM: 1) mineral-based, 2) plant-based, and 3) microorganism-based, which relate to the origins of the main raw material used. According to the natural materials definition in the Introduction section, mix designs mostly based on the use of fossil-based materials, mortar, concrete, glass, and alkali-activated materials (AAMs), sometimes referred to as *geopolymers*, were excluded from this review. This decision was made on the basis of the required high temperatures, raging between 1250 °C and 1450 °C, in order to process the constituent materials into additives applicable in construction [61,81,97], and therefore a kiln or furnace to be produced, and the material undergoes an irreversible transformation. PLA was also excluded from this review. For instance, even if PLA is often categorized as biological material because it originates from a renewal substance, namely corn, it has been criticized by some authors for its high environmental impacts and the fact that it is not naturally biodegradable but solely industrially compostable [70].

3.3.1. Mineral-based material family

The mineral-based family includes several specific sub-categories: earth-based [2,4,46,50,56,67,83,111], salt-based [43,50], sand-based [69,106] and ceramic-based [15,79] according to the main material used in the mix design. Fig. 4 connects these categorizations with the two identified AM categories, namely material extrusion (ME) and binder jetting (BJT). Further classification is identified for final printed objects that are burnt vs. raw (left to dry in air temperature). Burnt elements are identified to include mostly clay in an average temperature of 900C°, and are therefore termed “ceramics” [15,79] (Fig. 4 and Table 2).

Multiple papers in this material family discuss water content as a crucial element to take into consideration when reducing shrinkage and cracking while assuring good flowability and printability. For instance, Claire Im et al. [29] added alcohol to the water, and nylon fiber to the overall clay mixture, to create intricate spatial lattice structures. Shafiei et al. [103] mixed algae and clay to examine hygroscopic characteristics and interaction with water of the final product. Young et al. [112] developed their specific clay mixture by adding chamotte clay (also known as *grog*, a type of clay that has been fired at a high temperature and then crushed into granules or powder) as an additive to the clay material. Chamotte clay was shown to reduce the sintering stresses and reduce local cracks and flaws [36]. Furthermore, Young et al. [112] also added Dispex, a commonly used deflocculation industrial slip casting, to create maximum fluidity with the lowest possible volume of water (again, to reduce shrinkage during drying).

The review of the earth, clay, ceramic, salt, and sand-based papers shows that the focus of these papers is mostly related to geometrical investigations. In papers that focus on earth-based materials, Perrot et al. [91] demonstrate that material extruded using a rectangular cross-section nozzle exhibits higher compressive strength as opposed to cylindrical nozzles. Table 3 summarizes the different mix-designs developed for AM with natural and engineered soil. As a matter of fact, vernacular earthen construction methods rely on the characteristics of the soil found nearby. Adjustments can be made as needed by developing engineered soil with the incorporation of clay to enhance plasticity or sand to decrease it.

In terms of water content, earth-based mix designs in conventional (non-digital) wet construction, such as cob, require an approximate average of 24% initial water content [96]. Shown in Table 3, the water content in 3D printed earthen mixtures is similar to traditional earthen materials or often increased for 3D printable earthen mixtures, assumingly to achieve extrudability. Curth et al. [33] found that the shrinkage could be linearly reduced by reducing the water content while Ji et al. [67] also suggest the beneficial effect of increasing sand content for the same purpose.

Considering the use of natural soils and their inherent variability, characterization of the soil particle distribution is often shown and may be outmost importance to correlate with extrudability (given the flowability of the clay vs. the sand within the soil). Moreover, when using natural soils for 3D printing, the reviewed literature indicates a sifting process to remove large aggregate, stones, and other organic bodies, usually using a screening bucket with a grain size of 0–4.75 mm at the lab [18] or 1-6 mm on site [48,50,91]. Overall, it is shown that the amount of sand and other aggregates should be less than 85% to avoid frictional behavior and to ensure pumpability [91,100,111]. Rückrich et al. [100] evaluated the effect on compressive strength by changing different clay-to-sand volume ratios. Even if a 1:2 clay-to-sand ratio was characterized by a higher compressive strength, they found that, for printability, the mix designs required an increase of the amount of clay to 1:1 and 2:1 (called respectively in their manuscript $M1_{adjusted}$ and $M5_{adjusted}$). The choice of soil formulas can also be determined by the yield stress for printable mixtures [67]. In their research, Ji et al. [67] aimed at maximizing the sand content and reduce the water content to reduce the risk of shrinkage.

Overall, Table 3 highlights the large variability and discrepancy in defining the right clay-to-sand ratios and utilizing different units.

In most of these examples, small amounts of natural plant fibers (0.5 to 2 wt%), such as wheat straw and sisal, are added to prevent shrinkage upon drying [4,58,59,104]. The authors' previous work developed earthen blends incorporating natural fibers, spanning from the conventional “cob” (2%wt fiber) to a newly formulated “light fiber clay” (featuring up to 13%wt fiber content and constituting 49% by total volume) [2,18,22].

To prevent earthen materials from deteriorating and to enhance structural stability, some researchers propose the use of biopolymer additives [6,48,77]. Potato starch is used as an additive by Silva et al. [104]. However, they suggested that temperatures around 90 °C are required to prepare more soluble and homogeneous starch dispersions, thus questioning the upscaling potential on a construction site. Rice husk is also used as a bio stabilizer because of its high silica and low alumina content; however, an excess rice husk content can hinder the evaporation of water and reduce the compressive strength [48]. Sodium alginate addition was shown to increase the green strength [91] and plasticity [77]. Other biopolymers, such as methylcellulose, chitosan, locust bean gum, guar gum, and xanthan gum, were found to be effective for the same purpose [2,18,22]. More traditional, calcined additives are also used, such as lime-based binder hydraulic lime [4,48,58,59].

Printability tests in the reviewed papers show that the maximum stable height of an earth-based vertical “lift” (one freshly printed segment comprised of a series of layers) is 58 cm, which is very similar to

Fig. 4. Mineral-based research papers categorized according to the AM technique (material extrusion (ME) and binder jetting (BJT)) and the main material sub-category (earth-, clay-, ceramic., sand-, and salt-based).
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Table 2
Studies that focus on mineral-based materials in additive manufacturing, categorized according to the material sub-category and the main constituent substance (clay-rich soil, engineered soil, clay).

Mineral-Based Material Family		
Subcategory	*Main constituent*	*Reference*
Earth	Natural soil	[2]; [4]; [56]; [100]; [111]; [83]; [67]; [104]; [59]; [48]; [91]; [22]; [33]; [18]; [3]; [8]; [50]
	Engineered soil	[2]; [46]
Clay	Clay, left raw	[112]; [29]; [60]; [103]; [52]; [47]
Ceramic	Clay, burnt in kiln	[15]; [79]
Salt	Salt	[50]; [43]
Sand	Sand	[69]; [106]

the traditional cob method [58,59]. Moreover, Gomaa et al. [58,59] focused their attention on the most adequate pumping system to serve 3D printing with natural soil. As a result, they design a bespoke system combining screw jacks and a unique dual-cartridge design [58,59]. By leveraging conventional clay extrusion as a formal language, Friedman et al. [52] studied methods of woven deposition techniques to achieve diverse lattice patterning effects along the surface of the printing bed with a robotic arm. Farahbakhsh et al. [47] worked with adjustments to process parameters that have geometric effects on the clay material deposition process, resulting in varied patterns. Gürsoy [60] analyzed the geometrical variations of deposited clay using circular, elliptical, and wide nozzle geometries. This work proposed systematic variations in layer heights and nozzle heights, correlating the distance between the nozzle and the base plate and the resulted geometrical outcomes, as the extruded clay layers descended through the effect of gravity and are autonomously arranged into waves, curls, and sags with each successive layer.

Berman et al. [15] investigated gravity-stimulated printing design method for their ceramic coral-inspired prints. The nature-inspired hierarchical honeycomb ceramic structure evaluated by Maurath and Willenbacher [79] promises excellent mechanical strength at low density. In their mix design, they also added organic binders, such as palm wax, burnt during the kilning phase to result in assemblies with porous structures.

Existing 3D printing that utilizes salt is shown to incorporate BJT rather than ME technologies. Fratello and Rael [50] used one part by weight of maltodextrin and eight parts of finely ground powdered salt, and sake as binder. El-Mahdy et al. [43] incorporated traditional salt-based building mixtures inspired by Karshif, local Siwa desert earthen mixtures, while adjusting them to BJT by using proportions of salt, sand,

Table 3
Mix designs of the earth-based AM papers, in terms of soil, fiber, additive, and water content expressed in wt% (unless specified otherwise).

Natural soil				
Natural Soil [wt%]	Fiber [wt%]	Additives [wt%]	Water [wt%]	Reference
78–84% natural soil (composed of 4% medium sand, 30% fine sand, 41% silt sizes, and 25% clay)	1% sisal fibers, obtained from *Agave sisalana* cut in lengths of approximately 10 mm. The average diameter of the fibers was 137 μm, resulting in an aspect ratio (l/d) of 73	5% potato starch	16–22%	[104]
73% subsoil	2% straw	/	25%	[58,59]
67.6% subsoil	/	2% alginate	30.4%	[91]
72–73% subsoil	2% straw		25–26%	([56]; [58,59])
Filtered soil	/	hydrogel	/	[83]
30% w/v subsoil, 15%*w/v* silica sand, 22%w/v clay (with 1:1 ratio of China and ball clay)	15%w/v straw	/	18%w/v	[111]
79.4% natural soil (GU) (60% sand, 32%silt, 8% clay)	/	/	20.6%	[67]
Recommends using the jar test to identify a soil with less than 30% clay	No info	No info	No info	[50]
43–27% natural soil (10% clay content)	2–10% fiber (hemp, sisal, straw, kenaf, banana leaf)	2–7% biopolymers (methyl cellulose, sodium alginate, chitosan, locust bean gum, guar gum, and xanthan gum)	55–60%	[2]
3–63% natural soil (10% clay content)	4–13% fiber (straw)	2–5% biopolymers (alginate, methylcellulose)	31–79%	[18]
3–13% natural soil (10% clay content)	8–13% fiber (straw, hemp, kenaf and banana pseudostem)	5% biopolymers (alginate, methylcellulose, locust bean gum)	60–79%	[22]
82%soil (30% clay and 70% sand)	2% fiber (straw)	/	16%	[33]
Engineered soil				
Engineered Soil [wt%]	**Fiber [wt%]**	**Additives [wt%]**	**Water [wt%]**	Reference
46.22% natural soil (68.24% clay+silt, 28.8%% sand, 2.96% gravel), 18.78–22.78% sand	0.5% natural fibers, including jute, coconut, sisal and goat hair	8.25–11% hydraulic lime binder, 1.41% unaltered rice husk, 1.375%shredded rice husk, 1.375–2.75% marble waste dust, 2.75% municipal solid waste incinerator bottom ash, 8.25–11% cement	24–20%	[46]
70.42% natural soil (composed of 30% clay, 40% silt, and 30% sand), 18.78% silica sand	/	4.7% Lime-based binder, 4.69% Hydraulic lime, 1.4% rice husk	No info	[48]
38.53–52.63% clay, 11.11–17.73% sand	1.06–1.5% straws	7.08–11.63% lime, cement 2.40–2.48	24.19–31.91%	[4]
37.4–25.4% clay, 45.6–61.6% sand	0–0.05% cellulose microfiber	/	13–17%	[100]
73% engineered soil (49% sand, 34%silt, 17% clay)	Min 0.5% fiber (nim 2 cm length)	/	26.5%	[67]
83.2% engineered soil (34% clay and 65%sand)	/	/	16.8%	[8]
13 vol% Kaolinite and 50 vol% Ottawa sand	/	1 vol% methylcellulose	13 vol%	[2]

and Ethylene-vinyl acetate polymer binder of 2:1:0.5, with the binder making up to 14% of the total mixture. Kladeftira et al. [69] explored the possibilities and challenges of using natural sand within outdoor, on-site BJT settings. Specifically, they discovered that employing phenolic substance at 150 °C to sand in BJT increases resistance to breakage as opposed to a petroleum-based resin binder.

3.3.2. Bio-based family

Bio-based materials found in 3D printing are mostly divided between plant-based *fibers* and plant-based *biopolymers* (Fig. 5 and Table 4). In case of naturally-occurring and fully biodegradable 3D printable biopolymers, which do not require any source of heat upon extrusion, papers were found to include cellulose ([20]; [71]; [19,40]; [24]; [99]; [37,70]), xanthan gum [1], soy protein [74], tapioca starch [95], corn starch ([49]; [72]), chitin/chitosan [39,114], agar [75], and lignocellulosic fibers ([98]; [54]; [38]).

3D printing using natural plant fibers is shown to mainly utilize finely-shredded plant fiber, and very few attempts are shown to incorporate long plant fiber in 3D printed materials. The LivMatS pavilion stands out as one of the limited instances of, namely flax, with a coreless filament winding (CFW) fabrication technique [54]. This paper compares a fully biobased epoxy resin vs. a fossil-based one, deciding to use only the fossil-based one to optimize the structural design. Additionally, the use of hydrophilic fibers like flax poses impregnation challenges when combined with epoxy resin; addressing these issues requires adjustments at the fabrication level and further exploration of suitable fiber-resin combinations, complemented by impregnation tests.

Rech et al. [98] developed 3D printable mixtures with xanthan gum, which serves as a binder for cellulose insulation, and sawdust, along with a blend of bio-based plasticizers (glycerol) and water, for flow properties. They also explored various reinforcing fillers, demonstrating that a small incorporation of inorganic material (vermiculite) can notably enhance the mechanical properties of bio-based mix designs not only during printing, but also in its final form after drying. Additionally, the study illustrates that the introduction of calcium ions could amplify the binding effect of xanthan gum, and when combined with an increase in binder concentration, could enhance the early-age strength of the 3D printed material [98]. Choi and Yi [26] developed several material recipes with biopolymers and natural wastes, while documenting the numerous trials and errors in material ratios and compositions. Their study shows that cellulose and kaolin clays can serve as fundamental

Table 4
Research papers on 3D printed bio-based materials, categorized according to the main constituent substance.

Material Family	Subcategory	Main constituent	Reference
Bio-Based	Fiber	Lignocellulosic fibers	([98]; [54]; [38])
	Biopolymer	Cellulose	[20]; [71]; [40]; [19]; [24]; [99]; [70]; [37]; [26]; [39]
		Soy protein	[74]
		Corn starch	[49]; [72]
		Chitin/chitosan	[39]; [114]; [37]
		Xanthan gum	[1]; [26]
		Agar	[75]; [26]
		Other	[26]

Fig. 5. Research papers on 3D printed bio-based materials, categorized according to material extrusion (ME), and the main constituent (plant-based fiber and plant-based biopolymer).
Images from: Ref. [26,37,38,40]. Copyright Elsevier, 2024, used with permission. Images from: Ref, [20,49]; [19], picture by ©Tatiana Campos, used with permission. Ref. [71], picture by ©Dunia Abdullah Agha, used with permission. Ref. [60], picture by ©Benay Gürsoy, used with permission. Ref. [70], picture by ©Christoph Klemmt, used with permission. Ref. [75], picture by ©Gitit Linker, used with permission. Ref. [72], picture by ©M. Kretzer and S. Mostafavi, used with permission. Ref. [1], picture by ©Secil Afsar, used with permission. Ref. [99], picture by ©Gabriella Rossi, used with permission. All rights belong to the authors at CITA / Royal Danish Academy All rights belong to the authors. Images from: Ref, [54,98], open access papers under the CCBY license. Image from [39] (all the images are retrieved from https://oxman.com).

elements for maintaining morphological integrity, while their use of eggshell, charcoal, and chlorella is shown to slow down biodegradation and mitigate shrinkage caused by water evaporation.

Several examples explore cellulose-based pastes in additive manufacturing, focusing on their printability characteristics, and the potential of using cellulose derivatives from recycling paper wastes [20]; [71]; [19]; [24]. These studies all show that cellulose-based pastes mix designs heavily rely on the quantity of added water. Duro-Royo et al. [40] use cellulose microfiber to obtain volumetric composite in a flow-based fabrication process. To control the shrinkage once dried, Rossi et al. [99] added sensors between layers to track the curing of the 3D printed cellulose-biopolymer objects. Klemmt et al. [70] used components that are either biodegradable or biocompostable, occurring naturally in the environment.

From a circular economy perspective, some biopolymers are selected in these studies given their abundance in the region where the research takes place, or even globally, such as corn starch and soy protein ([49,74]; [72]).

Multi-material structures made from blends of chitosan and cellulose were shown to be fully biodegradable, lightweight, and capable of self-support [39,40]. Dritsas and Fernandez [37] developed a process using cellulose and chitin, to create a fungus-like adhesive material deposited using an extrusion-based method. Instead of relying on the growing properties of fungi, these materials are artificially created to have similar mycelium composition but without the controlled conditions required for a living organism. These materials could be used for rapid prototyping when the objects produced are often impermanent, such as furniture, but also concrete formworks. Other fiber-biopolymer composites are shows to exhibit critical features in 3D printing. Flax shive-based lightweight composites have been demonstrated to be 3D printed in a process where a paste mixture of plaster and/or quarry fines is used to impregnate the fibers, resulting in a composite that rapidly hardens (in less than 30 min) to form an insulation layer. This layer could potentially serve as support for the printed structural material in a multi-material printer system [38].

3.3.3. Living-based family

Fig. 6 and Table 5 group the identified research papers on 3D printing with living organisms according to the organism used, namely mycelium, bacteria, algae, and silkworms. Some of these papers include a base paste with clay, fibers, and biopolymers. However, these papers were grouped together in a separate material family in order to highlight the main feature in these material solutions: to sustain the growth of organisms as the main binder, or "glue".

3D printing with mycelium is shown to mainly use the *saprophytic* fungi strain, which typically feed on lignocellulosic fibers and can be printable with a rheology modifier. It is therefore observed, in this line of work, that the development of a paste that could serve both as an extrudable and feeding solution for fungal nutrient is key [105]. To be used in 3D printing, clay or other biopolymers, such as chitosan, are shown to be mainly added to the fibers as a binder or viscosifier, and to assure buildability. Prior to the growth of mycelium, the 3D printed fibrous substance required a "temporary" binding agent to ensure shape retention in the desired configuration [55].

In 3D printing with mycelium, the living mycelium spawns or liquid cultures are inserted before printing, without the ability to regulate, or more precisely, "program," the growth of the mycelium itself in response to design requirements [55]. Other, more manual inoculation strategies are shown to work well when "implanting" the mycelium spawns within the contour areas while printing [23]. In both cases, the base paste, as

Table 5
Research papers on 3D printed microorganism-based materials, categorized according to the material family, subcategory, and the main binder.

Material Family	Subcategory	Main binder	Reference
Microorganism-based	Fungi	Clay	[23]; [55]; [66]; [31]
		Biopolymer	105]; [17]; [24]; [35]; [84]
	Bacteria	Bacteria	[7]; [12]
	Algae	Microalgae	[32]
	Warm	Silkworm	[89]

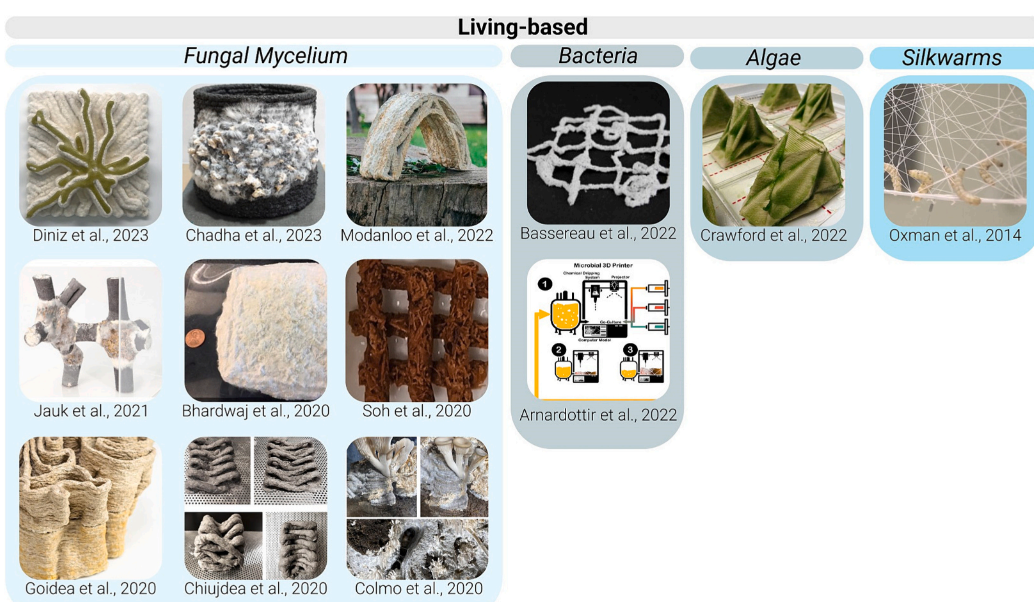

Fig. 6. Research papers on 3D printing with living-based materials, categorized according to the type of organism used (Mycelium, Bacteria, Algae, Silkworms). Images from: Ref. [17], Copyright Elsevier, 2024, used with permission. Images from: Ref. [35]. MYCO-ALGA by bioMATTERS. 3D printed mycelium and algae tiling system. Image provided courtesy of authors. © 2023 bioMATTERS, LLC. All Rights Reserved. Ref. [23], picture by ©Olga Beatrice Carcassi, used with permission. Ref. [24], picture by ©Ruxandra Stefania Chiujdea, used with permission. Ref. [66], picture by ©Julian Jauk, used with permission. Ref. [31], picture by ©Claudia Colmo, used with permission. Ref. [12], picture by ©Aurélie Mossé, used with permission. Ref. [7], picture by ©Martyn Dade-Robertson, used with permission. Ref. [32], picture by Assia Crawford, used with permission. All rights belong to the authors. Images from Ref. [55,84,89], open access papers under the CCBY license. Ref. [105], an open access article under the CCBY 4.0 license.

well as the mycelia strain, are key for both printability and inoculation performance. Strains such as the *Byssomerulius corium* and *Gloeophyllum* sp. are shown to 3D print well when mixed with sawdust chips, paper pulp, kaolin clay, and water to fabricate modular elements that form a spatial column [55]. Mix designs with mostly fibers and psyllium husk powder were shown to be fine enough to avoid fiber clogging at the nozzle level, while ensuring successful inoculation of the fungal mycelium [17]. Overall, it shown that mostly cellulosic components – such as recycled paper or cotton fibers – work best for this type of 3D printing. When mixed with water, a binding agent (biopolymer), a viscosifier (such as glycerin) and oftentimes additional nutrients (such as wood flour), such mix designs can obtain slurry consistency needed for 3D printing [24]. Other studies developed extrudable pastes by combining industrial and domestic materials often discarded, such as cardboard and sawdust, as mycelium substrates with algae and bacteria as natural dyeing [35]. Shredded paper was also shown to work well with natural gums as gelling agents, such as in the case with *Pleurotus Ostreatus* oyster mushroom and xantham gum [84]. The *Pleurotus Ostreatus* strain was also shown to be successful in a direct inoculation on top soil mixed with different biopolymers (i.e., xanthan gum, guar gum, water, wet hay, glycerol, molasses, perlite), for architectural potentials of mycoremediation (the phenomena in which fungi is used to degrade or isolate contaminants in soil) [98].

During the design and assembly of modular elements that are 3D printed with fungal mycelium, it is necessary to control shrinkage to ensure geometrical alignment within the final design. Volumetric shrinkage with 3D printed mycelium is reported to be up to 30% [55]. The structural capacity of mycelium-based is shown to range between 204.28 and 278.30 N [66]. Here, the mean tensile strength is shown to increase by 67% with *Pleurotus Ostreatus* mycelial growth, as opposed to the hardened mixture of clay and sawdust without mycelial growth [66].

Avoiding contamination was also shown to be key in 3D printed fungal mycelium; it is also shown that cleaning all machine parts that will be in contact with mycelium with a 70% ethyl alcohol solution is proven most helpful [17].

For 3D printing with other living, bacterial substances, a few demonstrations emerged in recent years. *Porosarcina Pasteurii* is one of the most efficient non-pathogenic bacteria used for microbially induced calcite precipitation (MICP). Different inoculation approaches were shown to be possible for this strain, including: 1) spraying the bacteria on the dry 3D printed nutrient substance, and 2) integrating the bacteria within the nutrient substance while still wet before printing. One prominent example is “Papier Plume”, a project that developed 3D printed nutrient foam derived from locally sourced paper waste, that can undergo 3D printing and be fortified through bacterially-induced calcite precipitation [12]. 3D printing with bacteria requires a secondary treatment with urea and calcium salt, which should be administered to trigger the nutrient foam’s calcification through urea hydrolysis. Other bacteria, such as the *Komogataeibacter xylinus*, can be used to produce bacterial cellulose following the printing process, as demonstrated by Arnardottir et al. [7].

Lastly, additional substances that are derived from the animal world, like silkworm, was shown to be a potent 3D printing material. For instance, the “Silk Pavillion” explores *Bombyx Mori* silkworm’s capacity to build a 3D cocoon from a single filament [89].

3.3.4. Additional additives

Despite their environmental and health-related benefits, natural materials often exhibit challenges in terms of their durability and structural capacities, which are lower than engineered materials such as petroleum- and cementitious-based materials. As a result, researchers are actively exploring the incorporation of stabilizing additives into their 3D printable mix designs to enhance mechanical properties.

Fig. 7 visualizes the identified additives and their use in 3D printed natural materials. As part of this review investigation, the analyzed papers were systematically cataloged according to the based material, the additive, and its function in the mix design, showing that the main use of additives is to reinforce or stabilize earth materials with biopolymers and lime.

Since a paste extrusion logic is used in all the material families categorized in this study, additives are fundamental in easing the material passage through the nozzle. One of the main constituents of typical 3D printable pastes is water. However, when the quantity the water is excessive, shrinkage and cracking increase. To reduce the water quantity, materials such as clay, chitosan, and cellulose are used to enhance the flowability, modify the rheology, act as thickening agents, and plasticize the mixtures to obtain more viscous materials for 3D printing applications.

Earth mix-designs are shown to be reinforced with both biopolymers such as sodium alginate, which was demonstrated to increase compressive strength by 7% [91] and faster structural build-up [57].

The second most used additive involves lignocellulosic fibers (categorized in the Bio-Based Material Family as *plant-based fibers* in this paper) used within earth, cellulose and mycelium mix designs to reduce cracking and improve ductility [48,49,58,59,76,104]. Natural fibers are also utilized as fillers and as nutriments for mycelium [23,66]. In mycelium-based examples, fibers are the main constituents, however, to assure extrudability and buildability materials such as clay and biopolymers must be added [55]. Gypsum and natural prompt cement can also be used as binders [38]. Prompt cement is siliceous cement formed in lower temperature ranges of 800–1200 °C, thus representing a more sustainable alternative to Portland cement. Moreover, quarry fines can also be used as a low plastic clay by washing concrete aggregates.

3.4. Properties of 3D printed natural materials

The reviewed research was cataloged according to a range of performance parameters, in order to identify the predominant performance analyzed in 3D printed natural materials, and identify missing areas of research. Shown in Table 6, the state-of-the-art literature was cataloged according to the following performance categories: material characterization, structural, thermal, environmental, printability, and geometrical performance.

A strong attention is shown to be paid to the characterization of raw materials, especially when natural soils are employed, assumingly due to their high variability. Particle size distribution, electron microscopy imaging, and X-ray diffraction (XRD) analyses are shown to be mostly used in the literature to address the variability of natural soils and clays. Structural investigations are analyzed to evaluate the mechanical behavior, especially compressive strength, of novel mix designs and to assess load-bearing capacity vs. infill possibilities, which is often conducted in the analyzed papers in comparison with traditional materials. Earth based materials are the most analyzed for their compressive strength with results vary between 0.87 MPa to 3.70 MPa [8,22,33,46,48,58,59,104]. The variation depends to the presence of fibers, stabilizers and nozzle geometry. The new *SaltBlock* demonstrated on the other hand a compressive strength of 9.5 MPa (relatively similar to old masonry Karshif) with a density of 1.4 g/cm^3 [43]. In contrast, concrete and fire bricks have a compressive strength of 24 MPa and 12 MPa respectively. Young’s modulus of around 200 MPa can be achieved for lignocellulosic waste fibers and particles mixtures by adding 2.6 wt% of xanthan gum and a 0.2 M concentration of calcium chloride [98]. The peak strength of silica sand, gypsum powder, and coated silica beads fabricated through binder jetting and selective laser curing were 6.21 MPa, 9.14 MPa, and 8.15 MPa, respectively [106]. Mixtures of chitosan with mycelium-enriched bamboo yielded a compression modulus of 40 kPa, compared to 240 kPa without chitosan [105]. In another study, oyster mycelium is grown on shredded paper, wheat bran, and guar gum to prepare a material that can be extruded and offers sufficient mechanical strength, namely a Young’s modulus 78.3 MPa [84].

In the case of natural fibers filament-wound composite structures, for instance, the experimental results are used to feed the computation

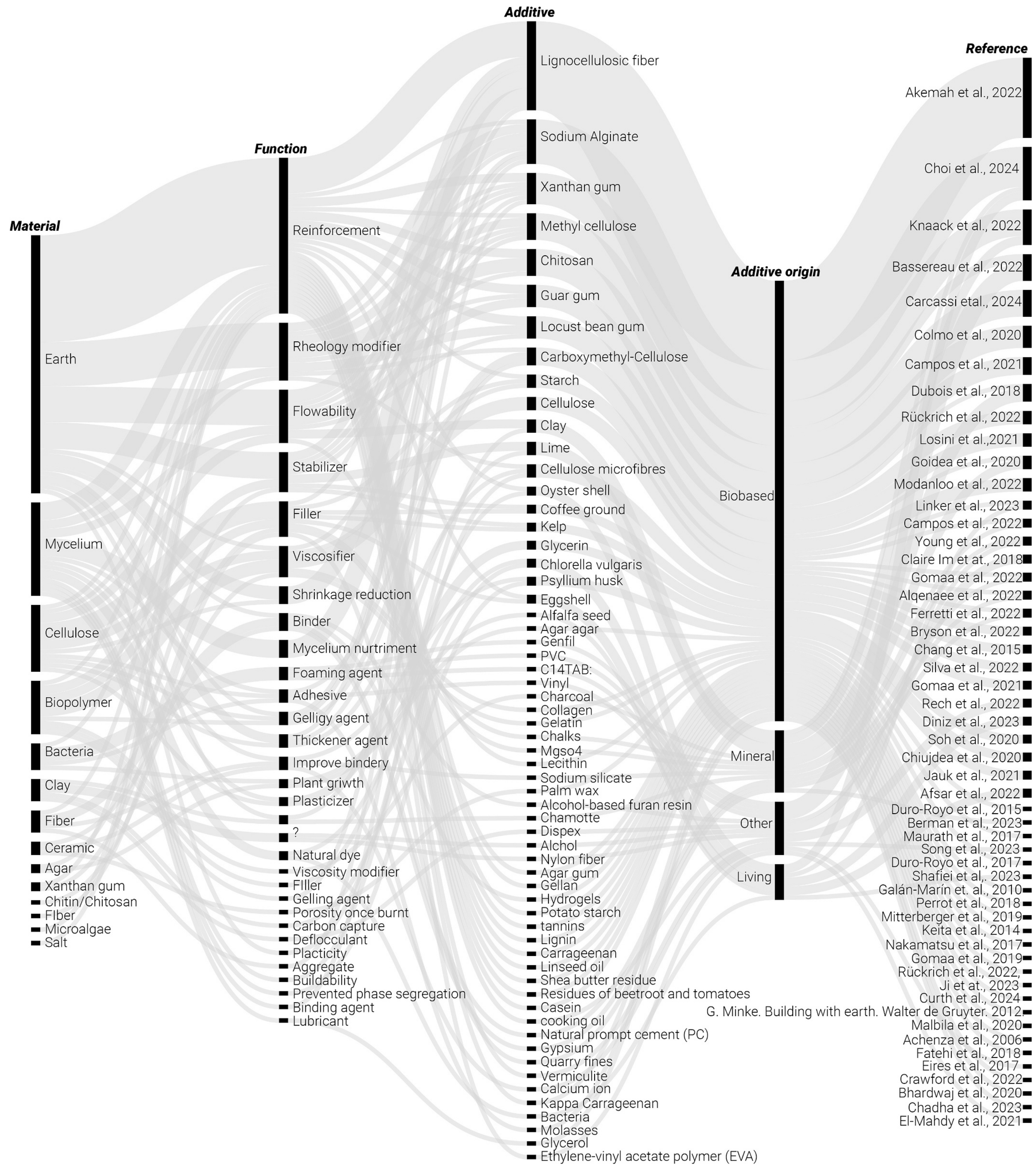

Fig. 7. Additives used in natural materials mix designs in AM, and the identified related functions served (data are available in Appendix A). Alluvial graph created with RAWGraphs [80].

software and simulate through Finite Element Model (FEM) the behavior of complex shapes that are only possible with the geometrical freedom provided by additive manufacturing [54]. Similarly, Gomaa et al. (2021) utilized Galapagos in the Rhino-Grasshopper package to optimize the wall cross section geometry in terms of printing path, nozzle size and wall depth and shell thickness to obtain a structural low-rise construction 3D-printed cob walls.

For environmental performance, several papers develop a lifecycle analysis (LCA) of the materials used, three of them [46]; [54]; [22] limited their analysis to only to the embodied carbon, while embodied energy analyzed only in one study [43]. A full-spectrum analysis from cradle-to-gate was also shown to be limited to one study [54]. The

Table 6
Properties of the 3D printed natural materials identified in the state-of-the-art.

Performance Category	*Subcategory*	*Reference*
Material characterization	Granularity testing and particles size distribution	[67]; [46]; [106]; [18]; [8]; [33]
	Microscopy (SEM)	[105]; [46]; [2]; [89]
	XRD	[8]; [91]
	Binding force between the printed layers	[66]
	Thermogravimetric analysis	[67]
	Porosity	[67]
	Mass Transfer Coefficient	[67]
	Jar (shake) test	[33]
Mechanical Performance	Compressive strength	[46]; [98]; [106]; [43]; [4]; [105]; [84]; Gil [54]; [18]; [8]; [58,59]; [38]; [22]; [33]
	Shrinkage	[46]; [103]; [98]; [12]; [33]
	Flexural Strength	[46]; [4]; [18]; [8]
	Rheology	[79]; [71]; [67]; [8]
	Tensile	[105]; [39]; [66]
	Plasticity	[4]; [8]
	Fall cone test	[67]; [100]
	Atterberg limits	[67]; [46]
	Splitting tensile	[4]
	Viscosity	[98]
	Flow table	[8]
	Finite element method simulation	[54]
	Workability (Vicat test)	[38]
Environmental Performance	From the cradle to the gate only GWP	[54]; [22]
	Embodied carbon binder	[46]
	Recipe Middle point	[50]
	Embodied energy	[43]
	Cradle-to-site	[3]
	Cradle- to- cradle	[33]
Geometrical Investigation	Pattern	[43]; [56]; [20]; [112]; [71]; [23]; [47]; [52]; [72]; [75]; [83]; [111]; [58,59]
	Curvature and continuous printing path	[19]; [52]; [75]
	Nozzle shape	[60]
	Monitoring of the curing process	[99]
	Multi-material system	[38]
	Gravity-Stimulated Printed Designs.	[15]
Thermal Performance	Thermal conductivity	[46]; [56]; [38]
	Thermal transmittance	[43]
	Specific Heat Capacity	[38]
	Thermal Simulation	[89]
Printing Parameters	Buildability and Extrudability	[4]; [71]; [100]; [17]; [19]; [1]; [70]; [67]; [2]; [18]; [8]
	Flow assessment	[40]; [47]; [100]; [8]
Other Performance Parameters	Acoustics	[38]
	Durability Performance (accelerated weathering)	[98]

environmental performance is mostly enumerated for binders, whereas the other constituents make a negligible contribution to CO_2 emissions when compared to the binder. The environmental analyses show the carbon emissions of 3Dprinted natural materials *binders* range between 0.052 $kgCO_{2e}$/kg for rice husk, marble dust, and lime, to 0.077 $kgCO_{2e}$/kg when cement is used [46]. The ReCiPe endpoint-score LCA method is used for materials like salt, showing a 80% reduction of environmental impacts as opposed to PLA [50]. For end-of-life phases, the full recyclability of earthen construction with a cradle to cradle approach was shown to result in a printable mixture after immersing the components for 24 h [33].

Additional research investigates the thermal performance of 3D-printed cob by focusing on a specific cob mixture and conducting tests with diverse geometric extrusions [56]. Employing a Heat Flow Meter, this study demonstrated that the precision facilitated by digital construction technology led to a substantial 23% reduction in thermal conductivity, attributable to the incorporation of air pockets (from 0.48 W/mk to 0.32 W/mk). Moreover, manually filling these air pockets with straw was observed to result in an additional 7% decrease in conductivity. The thermal transmittance (U-Value) is instead calculated for the SaltBLovk cavity wall prototype [43], which performed better (0.94 W/m^2K) than a traditional 12 cm-wide fire brick wall (2.45 W/m^2K).

Multiple studies aimed at characterizing printability of new mixtures. Qualitative analysis of extrudability (i.e., the characteristics of a material to be easily extruded from a nozzle) and a quantitative approach for buildability (i.e. number or consequent layers that the material can hold before collapsing) are shown to be used most ([4]; [71]; [1,2,17,19,67,70,100]. Others have devised processes relying on the utilization of diverse materials deposited through varying motion speeds, fluctuating pressures, and a range of water-based viscous material compositions [40]. Adjustments of printing parameters for specific geometric effects on the material deposition process are also shown to be used [47]. Digital image correlation of the in situ printing test can also been employed to assess the buildability and layer collapsing over the effect of gravity and the load of consequent layers [8]. A comprehensive checklist of characteristics that must be fulfilled to make a mixture printable, namely workability, flowability, pumpability, extrudability, buildability, shape retention, open-time, interlayer adhesion, and printability, is offered by Teixeira et al. [108].

Additive manufacturing enables new opportunities in terms of bespoke mix designs but also geometrical, pattern, and textural investigations. With a strategic material deposition, it is possible to enhance the thermal properties [43,56], and suggest the exact location for additional material deposition performance [23]. The optimization of the printing path, also called contour crafting, assures the creation of a continuous curve, which can take into account the difficulty of assuring the retraction of the material during nozzle travels and avoid possible delamination [19]; [52] [75]). Layer height and nozzle size/geometry are key parameters that can affect the final quality of the printed objects [60].

Upon drying, natural materials tend to evaporate their water content, hence it is fundamental to calculate the shrinkage intensity to be able to include it in the design phase. With this intent, movement sensors within the printed layers can be used [99], or, otherwise, a general rule of thumb can be shown for earth construction to be an approximate 15% shrinkage [82], while 30–35% for biopolymer pastes [37]. Even if most of the examples here collected work with a 3-axes deposition logic, other studies utilized the spatial freedom provided by robotic arms to create lattices [24]; [29]; [54]. The mix designs developed to this end tend to be more viscous and have a lower water content to retain the spatial shape, sometimes even without the support of a base layer. In other cases, the effect of gravity on filament deposition is intentionally used to generate gravity-simulated printed objects [15].

4. Conclusions

In the past decades, emerging research has been established in the building design and construction fields to advance additive manufacturing (AM) using low-carbon and socially-equitable natural materials. To investigate and analyze the state-of-the-art in this trend, this paper presents a systematic review that sheds light on the techniques, mix design ingredients, additives, and performance parameters of natural materials in AM. Three main material families are identified and thoroughly analyzed: mineral-based, bio-based, and living-based. Consequently, this review aims to offer a comprehensive overview of existing studies on AM and natural materials, intending to guide new research initiatives and identify potential future trends in the field.

In the literature review, 80 papers were analyzed. The findings were grouped per natural material families, AM technology used, mix designs and types of performances. Fig. 1 illustrates the growing interest in the topic since 2017, following the studies on robotically extruded clay and Oxman's research on biological materials and advanced technologies. Table 2 highlights that the journal "Construction and Building Materials" predominantly publishes on this topic, with eCAADe, ICSA, CAADRIA and ACADIA recognized as high-level conferences in the field. Fig. 3 indicates that Material Extrusion without heat is the preferred method for working with these materials. Figs. 4, 5, and 6 help visualize the current state of the art and the design appearance/scale of these materials. Table 3 emphasizes the two different approaches to printing with earth materials: the use of natural soil versus engineered soil. Additionally, it highlights the variability in proportions of sand, clay, and water. These discrepancies in the literature are likely due to the intrinsic variability of natural soil and the different types of clay minerals that can be found. Therefore, when developing new earth materials, it is crucial to compare studies using similar types of soil by performing particle size distribution, electron microscopy imaging, and X-ray diffraction (XRD) analysis. When working with bio-based materials, fiber length and durability issues are still open topics that required further attention. In the case of living materials, only a limited number of organisms were utilized in this particular field; therefore, in the future, unexplored species of bacteria, mycelium and others could potentially open new avenues in hybrid living/non living combination of manufacturing methods.

In 3D printing, achieving the right extrudable and buildable mix design is essential for a successful process. Fig. 7 allows for navigation through the different constituents used for various material families and their respective functions showing that typically additives are used to reinforce natural material mix design. Table 6 provides guidance on the analyzed performances in terms of material characterization, structural, thermal, environmental, printability, and geometrical aspects.

Emergent trends in the literature show that very recent and, presumably, future research is to focus on the structural, environmental, durability, material characterization and lack of codes and standards that could potentially facilitate the spread use of natural materials in construction. The analyzed literature shows that the majority of research efforts are focused on the geometrical or printability potential of earth- and biopolymer-based mixtures. However, no specific attention was dedicated to combined natural materials and bio-inspired geometries. The performance aspects of these elements, particularly their structural, durability, and shrinkage properties, show a strong tendency to focus on the material characterization and compressive strength of printed mix designs. Table 6 highlights that there is a lack of codes and standards for researchers to compare material properties, printing settings, or performance measurements which often rely on conventional techniques ones. This suggests a need to create a generalized database to facilitate and promote the use and spread of these material.

This review aims to provide readers with a comprehensive overview of the current knowledge on natural materials for additive manufacturing while highlighting recommendation for future studies. In particular, further research should focus on correlating material properties with specific applications and identifying appropriate architectural uses. Durability remains a critical challenge as the scale of application increases. Therefore, bio-inspired design methodologies and natural coating systems, such as oils, require further investigation. A shift in maintenance practices is also essential, as incorporating natural materials necessitates an acceptance of natural cycles and timelines. The use of materials that are naturally degradable within a human timescale is crucial for the sustainable use of planetary resources. Promoting a culture of care and repair for these materials is preferable than relying on synthetic, long-lasting materials that contribute to increase the landfill waste. This approach not only respects the environment but also fosters a more sustainable relationship with the materials used as well.

Moreover, the availability and sharing of data regarding material ingredients, when extended to encompass a broader analysis beyond construction materials alone, could significantly contribute to the advancement of innovative mix designs. Examples of locally sourced materials and reinterpretation of traditional techniques suggest [43] an interesting way to enlarge the combination of AM with human crafts and local materiality. However, an in-depth exploration of environmental and social life cycle assessment details to validate the often-only assumed hypothesis of their positive impact and the consequences of including a machinery vs human labor is still missing in the literature.

One of the limitation of the study is that the analysis started without a definition of natural materials a priori. In fact during the cataloging phase, the authors had to question the very definition of "natural material," and as a result, it became possible to propose a tailored definition. In conclusion, natural materials are here defined as *locally available* from biological, living and geological resources, *low-carbon*, *recyclable*, and/or *safely disposable*. Moreover, they need to be *transformable* or *minimally processed* through *accessible technologies* that could be used in and near the construction site to lower the distance between the production and the fabrication locations. In future analysis, the concept of origin should not solely focus on the biological or geological sphere, but rather expanded to other readily available waste materials coming from the Technosphere. Moreover, considering durability performance, maintenance requirements should be key during the conceptual and construction phase. Another limitation of this work is the small size of the corpus, namely 80 papers. Here the focus was on scientific contributions, while a lot of the work on natural materials for AM as been developing in not research oriented milieux, such as makers collectives and FabLab's and in experimental designers' practices that use other means of disseminating their work.

When it comes to minimally processed materials from biological and geological resources, one can be forced to face and incorporate a certain degree of material variability. Simple and "low-tech" fabrication processes should be developed to harness local material availability, traditional practices, and opportunities for emergence in digital fabrication. Adopting the Ruskinian concept of savageness, this review conforms to Dickey's proposal for future development, encouraging a departure from technology's inclination to produce uniform outcomes. Instead, this work embraces the notion that qualitative characteristics emerge from imperfections, including material irregularities introduced by machines and natural material variation [34]. The ultimate goal of this work is to catalyze 3D printing using natural materials in large-scale building scale for the outdoors. So far, the scale remains at the material or component/pavilion level, with little to no examples at the architectural scale. Future applied research should focus on large-scale development, testing for weather resistance, and assessing the aging behavior of 3D-printed objects in commercially available systems.

The presented work asks to deepen knowledge in order to inform building standards, machinery development, and design demonstrations that utilize sustainable and just material practices for all, informed by equitable decision-making when planning future testing campaigns.

CRediT authorship contribution statement

Olga Beatrice Carcassi: Writing – review & editing, Writing – original draft, Visualization, Validation, Methodology, Formal analysis, Data curation, Conceptualization. **Lola Ben-Alon:** Writing – review & editing, Validation, Supervision, Funding acquisition, Formal analysis, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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Appendix A. Supplementary data

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