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Building a Greener Future: Advancing Concrete Production Sustainability and the Thermal Properties of 3D-Printed Mortars

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Abstract: The integration of waste materials in extrudable cement mixtures has the potential to make the construction industry more sustainable by reducing carbon footprints and developing eco-friendly materials. This along with advancements in 3D concrete printing (3DCP) provides engineering and architectural benefits by reducing material waste and costs. In this paper, the impact of waste incorporation on properties of mortar and concrete is examined. The use of waste materials, such as pumice, coal slag, agricultural lignocellulosic residues, and recycled rubber tyres, to improve thermal insulation and durability of cementitious composites is discussed. In addition, the incorporation of air-entraining admixtures with surfactant activity is explored for their indirect effect on thermal behaviour, pore size reduction, and enhancement in concrete properties. This review includes important topics such as a strength resistance to freezing and thawing, fire resistance, plasticising effect, and delay in cement hydration. These findings highlight the benefits of using diverse waste materials in construction, providing a multidimensional approach to waste management, cost optimization, and enhanced construction materials in the context of 3DCP.

Keywords: 3D concrete printing; waste; mechanical properties; thermal insulation; admixtures



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1. Introduction

In the context of the application of circular economy (CE) principles [1], the European adoption of the Green Deal presents a joint effort in the implementation of strategies toward a competitive and efficient utilisation of resources' economy that minimises environmental footprints, thus envisioning a carbon-neutral Europe in the near future [2].

The building and construction (B&C) sector is considered one of the key sectors for CE adoption due to its massive impact on resource consumption, waste generation, and environmental emissions [3,4]. As a result, this sector has become the focus of developing new policies and launching a new comprehensive strategy for a sustainable built environment [5,6]. Understanding the environmental impact of the B&C sector and cement manufacturing is becoming increasingly important since concrete production is responsible for 9% of total emissions of greenhouse gases (GHGs), known to exert an accentuated effect on climate change, with about 0.8 tons of CO₂/ton of cement resulting from operations of transport, mixing, and application [7]. Improved production methods and formulations that reduce or eliminate CO₂ emissions from the cement manufacturing process are mandatory, as well as to counteract the impacts on the product cost of new regulations, green taxes, and escalating fuel prices [8].

A relevant perspective to consider is reducing the high amount of energy consumed by the cooling and heating of buildings, estimated to account for approximately 40% of primary energy and 36% of greenhouse emissions in European Union member states [9].

This fact, and the demand for housing due to the expected growth in the human population of about 10.9 billion in 2100 [10], is arousing many concerns about the negative impact on energy and climate goals [11]. One solution to increase sustainability and minimise the related environmental footprint of B&C is incorporating waste in concrete production as supplementary cementitious materials (SCMs) or fillers from several sources (construction and demolition, industrial, agricultural, etc.). This action will contribute to proper waste management and can fundamentally improve the thermal properties of cementitious composites, resulting in significant energy savings, correlated costs, and lower GHG emissions [12,13].

Since the beginning of this century, the application of additive manufacturing methods such as three-dimensional (3D) printing to concrete production and the fusion with the Contour Crafting technique [14] has gained ground over the established procedures of construction [15]. The computer-controlled deposition of extruded mortars layer-by-layer [16], also called 3DCP, coupled with Building Information Modelling (BIM), is already considered a practical tool mainly used for the fabrication of both structural and non-structural concrete elements [17]. While many advantages are already recognised, the complete adhesion of the sector to this technology depends on legislation that supports the pioneer companies since the investment has been focused mainly on the necessary automation and the demand for skilled labour, neglecting the sector's economic, social, and environmental benefits [18].

This paper aims to summarise and discuss the strategies followed so far concerning the integration of wastes in the design stage of cementitious composites to enhance the mechanical and/or thermal performances in the building infrastructures, integrating digital construction as a new path to CE.

The survey of the relevant literature was found within the article, title, abstract, and keywords. The keywords used for the systematic search are "cementitious mortars", "waste integration", "admixtures", "additive manufacturing", "3D concrete printing", "mechanical properties", and "thermal insulation", and priority was given to scientific articles, also paying attention to their number of citations. Thus, a good majority of the articles collected are in journals indexed in ScienceDirect (such as *Construction and Building Materials; Journal of Cleaner Production; Journal of Building Engineering; Cement and Concrete Composites; Automation in Construction;* and Engineering Structures) and Scopus (such as Buildings; Materials; Applied Sciences; and Sustainability).

2. Three-Dimensional Concrete Printing Prerequisites

The 3DCP technology allows rather complex and customised design, providing a quick construction method on and off site without precast, reducing the workforce, optimising material utilisation, and reducing materials waste, construction costs, and carbon footprints [11,19]. In opposition to conventional construction methods, 3D printing (3DP) is a highly customisable technology that allows the integration of nature-based infrastructures in the design stage, thus enabling the incorporation of secondary/recyclable materials with low environmental impact [20,21], providing a solid construction system based on robotic-assisted extrusion that is necessary to ensure the continuity of the printing process, a maximum extrusion rate, the consistency and quality of layers, freedom of movement, and reduction in human interaction [22].

Figure 1 shows the equipment of a gantry system for 3DCP.

Factors related to the mortar formulation and the printing process, such as the printing speed and mobility, flow rate, pump pressure, layer height and width, nozzle geometry, and environmental conditions, play crucial roles in affecting concrete performance in both fresh and hardened states [15,23].

Given its known properties, versatility, and ease of application, the choice for cementitious mortars on a global level is quite immediate. The formulation of cementitious mixtures proposed by different authors can vary considerably along with the printing system, the material supplier, and the equipment operator [24]. Table 1 displays the charac-

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teristics that all designed mixtures must present [25–27] with some relevant references to experimental research.



Figure 1. Gantry system constitution for 3DCP: (a) 3D printer; (b) mixer; (c) pumping system; (d) extruder with nozzle (from Be More 3D).

Table 1. Printable mortar's technical demands.

Properties	Description	
Flowability	Transportation of the paste from the mixer to the extruder	[28,29]
Pumpability	The ability of the material to be extruded without phase separation under pressure	[30,31]
Open time	Maintenance of the material's consistency for good extrudability	[32,33]
Extrudability	Extrusion of the material in continuous and uniform filaments	
Buildability	Extrusion of the mortar in stacked layers and retention of the extruded shape	[34,35]
Mechanical strength	Compressive and flexural strength	[36,37]

Mortar's rheology and particle-size-dependent yield stress [38,39] are fundamental parameters that directly influence the mortar extrudability and the integrity of the bottom layers to meet stack layer-by-layer requirements and a large height-to-weight ratio typical of 3DCP of buildings [40–43]. Each layer must be able to support the weight of the subsequent ones so there are no cracks or collapse of the element.

If the size of a mortar's aggregate is incompatible with the nozzle diameter, it will lead to additional pressure, inducing clogging problems in the pumping system [44]. This will impact the extrusion rate, consistency, and printing velocity, leading to excessive layer porosity, poor interlayer bonding, and weak mechanical properties derived from geometrical imperfections and surface shrinking. This will culminate in failure due to elastic buckling and plastic collapse [45,46]. Therefore, to satisfy the required fresh properties of printable concrete, such as pumpability and extrudability, the content of a coarse aggregate needs to be significantly reduced, leading to a quantity of OPC in the mixture design typically 40% higher than the conventional concrete [47,48] and a higher cost of the concrete material [41].

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To achieve economic and environmental sustainability, it is only logical to employ waste materials to replace OPC or as a part of a fine aggregate in mortar, solving two additional problems: the shortage of natural sand and the massive increase in waste disposal [49].

Chemical additives incorporated as admixtures in the mortar (e.g., superplasticisers, setting accelerators/retarders, and viscosity-modifying agents) will manipulate the mortar's rheology, maintaining the workability and buildability over time, hence controlling the yield stress evolution [50,51].

A panoramic view of waste availability is essential to evaluate the feasibility of waste integration in concrete mortars.

3. Waste Availability

Consulting the report provided by Eurostat [52], the production of waste generated by activities in 2018 for the 27 members of the European Union (EU27) shows that construction (35.9%), mining, and quarrying (26.6%) are the activities that produce the highest amounts of waste, followed by manufacturing (10.6%) and waste/water activities (9.8%) and Agriculture, Forestry, and Fishing (0.9%).

In recent decades, the massive destruction of old structures has been the source of enormous construction and demolition waste (CDW). In Europe, the generation of this class of debris is estimated to be around 820 million tons/year, representing almost 50% of the total waste in the sector [52,53]. Moreover, EPA pointed out a CDW production of 600 million tons in 2018 in the USA, where concrete was the most significant portion at 67.5%, followed by asphalt concrete at 17.8%, more than twice the amount of generated municipal solid waste [54].

Several initiatives to promote circularity include CDW waste as a central aspect [55], and reusing them in concrete as a substitute for virgin aggregates is considered efficient [56].

Mining and processing industries are among the most intensive sources of environmental pollution, with wastes making up over 90% of raw materials extracted from minerals [57]. Managing such large quantities and the resulting long-term accumulation (estimative of 19 billion solid tailings until 2025) constitutes the most significant waste problem on the planet [58]. The waste generated by mineral extraction is waste rock, sludges, and non-metallic (e.g., pumice, sand, quartz, phosphate, or forsterite) or metallic (copper, iron, zinc, lead, etc.) tailings. The vegetation and overburden may also be considered waste [59]. An estimated value for the worldwide production rate is 35×10^9 tons per year [60]. Slags are by-products of the metallurgical smelting of ores and used metals [61].

The pressure on natural sand extraction makes mine waste and metal tailings a potential replacement for natural fine aggregates [62,63] and a way of avoiding the major environmental problems due to soil contamination and the infiltration of surface and groundwater with heavy metals and processing chemicals. To address the sustainability concerns, a significant strategy is to mobilise those industrial by-products [64,65] to reduce the impact on carbon footprints.

Manufacturing by-products is the most extensive category covering discarded material during or after processing. Some examples are waste from the processing of mine ores or wastewater treatment, oils and fatty acids from oil refining plants, sawdust produced by the furniture industry, cellulose from paper production, and plastics.

More available leftovers from manufacturing processes or municipal solid wastes are artificial fibre wastes, glass fibre, polypropylene, and polyester, and rubbers are also available [66,67]. These materials have a high potential for environmental hazards due to their lower biodegradability and the probability of leaking into the ocean or other public spaces [68]. In 2015, the primary plastic production amounted to 407 million tons, with around 302 million tons (75%) ending as waste [69]. Despite ongoing management systems that lead to their growing recyclability, plastics must still be incinerated and landfilled [70].

Sludges from wastewater treatment plants containing heavy metals, like aluminium or other toxic residues [71], need previous solidification and stabilisation to be included in concrete [72].

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Agro-wastes are generated mainly from forestry activities or the harvesting and processing of raw agricultural products, such as crops, fruits, poultry, dairy products, etc. [73]. The most produced agro-food wastes are rice/corn husk, sugarcane straw and bagasse, barley husk, eggshell, coconut, peanut shell, cotton stalks, and wheat straw residues. These wastes find direct uses in biofertilisers, mud houses, animal feed, biofuel, and heat generation in small-scale industries [74].

Eco-friendly materials designed for building applications seek to incorporate more agricultural waste and other cellulose-derived materials into mortars. This kind of waste usually offers a high availability, low cost, versatility, low weight, low energy consumption, and easy processability [75–77].

3.1. Impact of Waste Addition on Concrete Performance—The Current Situation

Driven by the increasing demand for sustainable construction solutions, the utilisation of waste materials in concrete production is a prominent focus in recent literature reviews. Various waste materials, including fly ash, silica fume, ground granular blast-furnace slag (GGBFS), and others, are being explored as partial replacements for traditional concrete ingredients. These materials contribute to enhanced mechanical performance, durability, and environmental sustainability of concrete structures. Additionally, waste materials such as clay brick powder, waste glass, ceramic powders, and tailings are being investigated for their potential to replace natural aggregates or cement in concrete mixes. Wastes derived from biomass such as wood, rice husks, bagasse, plant fibres, and leaves, among others, provide a structural advantage in cementitious mixtures due to increased specific properties (ratio of strength to density). In addition to their lightweight nature, recycled polymers incorporated into the mixtures offer advantages in terms of water absorption and corrosion resistance, and, in some cases, they improve the processability of the mortars.

Incorporating these waste materials reduces environmental impact and addresses economic and availability concerns associated with traditional materials. Strategies such as fibre reinforcement further improve the mechanical properties of concrete, particularly in printable materials used in 3D concrete printing (3DCP). Ongoing research aims to optimise waste utilisation, improve concrete properties, and advance sustainable construction practices.

The growing demand for concrete building blocks is satisfied with the partial replacement of waste materials from various industries.

After consulting recent literature reviews regarding conservative formwork construction, it became apparent that most of the authors use a combination of cement, sand, fly ash, silica fumes, ground blasted-furnace slag (GGBFS or GGBS) and other slags (e.g., copper, lead–zinc slags), and micro-fibres [13,78]. The same strategy is still employed in mortar composition using the 3DP technology, and the experimental approach was the subject of a recent and detailed review by Dey et al. [79].

Fly ash (FA) is a by-product from coal production and combustion that usually replaces OPC in percentages between 15% and 30% [80].

Silica fume (SF) is the powdered by-product from producing elemental silicon or alloys containing silicon in electric arc furnaces with a high content of amorphous silicon dioxide [81]. In ultra-high-performance concretes, SF is usually present in elevated percentages (10 to 30%), inducing densely packed microstructures with reduced porosity [47,82–84].

GGBS is obtained by quenching molten iron slag and mainly consists of a silicate and aluminosilicate of calcium and can partially replace clinker in cement by up to 50% [85].

FA, SF, and GGBS additions in concrete lead to early heat production due to a slower hydration reaction preventing thermal cracking, increasing resistance to a chemical attack, and improving mechanical performance, abrasion resistance, and long-term durability of the built structures [86–90]. Excellent environmental performances are achieved with a multi-component binder mixture with FA, SF, or/and GGBS or FA and SF in the formulation of printable mortars [47,91].

Those by-products and natural pozzolanic materials, such as calcined clay or metakaolin, can partially replace the binder in concrete production. Although they do not display

cementation properties on their own, when finely divided and in the presence of moisture, the silica and alumina contents react with the calcium oxide or calcium hydroxide present in the cement [92]. This reaction affects the mortar's rheology and the cement hydration process, improving overall mechanical properties [39].

The economic viability of green concrete production depends highly on the waste accessibility/proximity, generated amount, and chemical and physical consistency [82]. Fly ash's long-term global availability is compromised due to the increasing demand for concrete and the replacement of coal-based power plants with natural-gas-based power plants [79].

Several strategies have been employed to overcome this issue. Recovering ashes and other natural pozzolans in landfills and ponds presents a potential solution [86]. By-products such as biomass ashes (agro-waste and forestry) or ashes from other unconventional sources are good alternatives.

Table 2 references some examples, contemplating both the casting process and 3DCP. Figure 2 shows an example of the use of sugarcane bagasse ash (SCBA) in 3D-printable mixtures.

Source	Reference	ces
Biomass-Based	Casting Process	3DCP
Wood (power plants)	[93,94]	-
Rice husk	[95,96]	[97,98]
Sugarcane bagasse	[99]	[100]
Bamboo leaf	[101]	-
Other		
Palm oil fuel (POFA)	[102]	-
Municipal/industrial waste (solids and sludges)	=	[103]

Table 2. Alternative sources of ashes (pozzolans).



Figure 2. A 3D-printed element with a mixture based on SCBA.

Since fillers and tailings are the waste categories with substantial representation [79], clay brick powder, waste glass, and ceramic powders (e.g., table and sanitary ware, electric

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insulators, floor, tiles, etc.) can replace natural coarse aggregates or cement, attaining suitable strength [104–106].

Glass powder, up to 15% in concentration, showed significant improvement in the properties of end-cast concrete [105,106]. Different gradations of recycled glass in printable mortars influence porosity, pore morphology, and related crack propagation while obtaining opposite effects on flexural properties in different loading directions [107].

The presence of granite, ceramic filler, or siliceous fillers with powder-size particles in cast concrete reduced the mechanical performance while increasing the absorption area, accelerating the hydration process, affecting the mortar's drying shrinking performance, and improving adhesive strength [108].

Concerns due to higher water absorption raise problems that affect consistency and plasticity, leading to workability issues [109]. The size, content of particles, and percentage of waste replacement have some influence on the fresh and hardened properties of concrete. In addition, durability is one of the significant issues since recycled concrete structures are exposed to carbonation-induced corrosion. Carbonation depth in concretes with a 25 or 50% recycled aggregate was 1.07–1.20 times greater than in conventional concretes with a 100% natural aggregate [110].

Meanwhile, mixing fly ash (FA) and irregular limestone aggregate micro-fines (AMFs) with concentrations up to 12% in the mortar formulation displayed a synergistic effect. Despite reducing flowability and extrudability in the fresh state, AMFs improved the hydration reaction speed with increased shape stability and long-term strength [111]. The relation between printability and mechanical performance with size gradation and interlayer distribution was proved by incorporating recycled concrete, desert sand, and river sediment [112] and preserving buildability with up to 50% replacement of sand with spent garnet [113].

Tailings consisting mainly of silica, aluminium, calcium, and iron oxides are used in cement clinker production to increase mechanical performance [114]. Casting concretes with tailings added as fine aggregates from lead–zinc [115] and gold [116] ores led to increased strength in compression. Printable mortars incorporating copper and iron tailings revealed a close relationship between fluidity and setting time and material concentration and the relation of particle size with water absorption [24,117].

Concrete with iron and aluminium fillings and GGBS exhibited better mechanical performance (compressive, tensile, and flexural strengths) and durability properties, creating bonds between fibres and cement mortar across the cracks that promoted a superior resistance to crack propagation [118,119].

Two detailed reviews concerning slag valorisation concluded that copper slag improves the concrete chemical resistance to corrosion and carbonation/freezing—thawing resistance with an optimum mechanical performance obtained with 40% replacement and particle sizes below 10 mm [120] from the mechanical properties' perspective; lead and zinc slags could be used in mortar and concrete mixes for up to 50% replacement [121]. A study concluded that the water absorption rate and mechanical and durability characteristics of a concrete mixture incorporating fly ash (15%), aluminium dross (10%), and quarry dust (20%) are better than those of standard concrete [122].

Printable materials like in conventional concrete casting processes require reinforcement to counteract their low tensile strength [123]. The addition of fibres such as polypropylene, nylon, steel, copper, basalt, and glass fibres improves resistance to cracking and tensile strength and flexural capacity [24] and is efficacious in improving the explosive spalling resistance and residual mechanical properties in the case of fire [123–127], positively impacting printing speed, setting time, and yield stress [103].

Table 3 summarises the waste utilisation regarding 3DCP with referenced articles and assesses the impact on specific fundamental properties, such as the pumpability, flowability, buildability, viscosity, interlayer bond, shrinkage, and more.

Table 3. Examples of the impact of waste additives in 3DCP mixture properties (summary).

Waste	Optimum Replacement	Material Replaced or Reinforced	Concrete Properties	Reference
Magnesium oxide from waste brine	3%	Cement	Good extrudability, flowability, and buildability, ↓ sorptivity	[128]
Fly ash (FA) and silica fume (SF)	60:10%	Magnesium potassium phosphate cement	Variations on setting time, with good extrudability and buildability	[39]
Fly ash (FA) and granulated blast-furnace slag (S)	0:60%	PC	The superior material efficiency index	[47]
Rice husk ash	20%	PC	↑ Workability than control	[97]
Municipal sludge waste ashes (fly and bottom) and nylon fibres (NF)	7.5–10% FA 0.14% binder weight	PC	↑ Buildability and yield stress ↓ Setting time, flowability, and interlayer bond	[103]
Recycled concrete (RA), ceramsite particles (CPs), and desert sand (DS)	Depending on particle size and distribution	Aggregates	Adequate flowability and extrudability ↑ Interlayer bond and can ↑ shrinkage	[112]
Wood chips from spruce with different binders	Chips/cement = 0.15 Water/cement = 0.80	Aggregates	Density = 0.7 to 0.8 g/cm ³ Mechanical strength is enough for non-structural applications	[129]
Stone sludge + Al-polishing waste + cork + eucalyptus ash + superplasticiser SIKA	2:1 cement/stone sludge with superplasticiser Sika control 40	Composite with concrete	Good flowability, extrudability, shape retention, buildability, and open time Conventional mortar ↑ early age strength development properties	[130]
Nano-attapulgite clay	0.5%	Plasticizer	Compared with the control mixture † Viscosity recovery and structural build-up	[131]
Cellulose fibre (CS) and limestone, silica fume (SF)	Ratio—0.15 HB-CSA: 0.85 OPC: 0.15 SF	Filler for application between layers	↑ Printing interval, interlayer strength, and durability ↓ Voids and longitudinal flaws	[132]
Copper tailings	30%	Sand	Favourable buildability and high mechanical strength	[24]
Copper and iron tailings	Mass ratio—1:4	Water treated	↑ Mechanical properties	[117]
Recycled glass	10%	Sand	↓ Mechanical properties ↑ Flowability	[133]
Thermally treated peat-based admixture (MT-600)	0.5% of cement fraction [0.08–0.125] mm	Cement	↑ Strength of hardened cement pastes at an early age (3 days) than reference composition	[134]
Polypropylene fibres	Lengths 3 (M3) and 6 mm (M6), 0.1 to 0.3% v/v	Aggregates	↑ Workability ↑ Porosity (in the hardened state) ↑ Flexural strength (M6) ↓ Total free shrinkage (M3)	[123]
FA, SF, and ground granulated blast-furnace slag (GGBS)	20 wt.% (FA), 15 wt.% (SF), and 10 wt.% (GGBS)	Cement	↑ Compressive strength ↓ Flexural strength	[91]
Limestone aggregate micro-fines (AMFs)	3% < AMF < 12%	FA	↓ Flowability ↓ Extrudability ↑ Shape stability ↑ Green strength ↑ Compressive strength ↑ Flexural strength (after 60 days)	[111]
Spent garnet (SP)	SP ≤ 50%	Natural sand	↓ Green strength ↓ Young's modulus -Buildability	[113]
Recycled sand (RS) (crushed from waste concrete)	25 and 50% (only tested values)	Natural sand	Change from plastic to solid material It affects early-age behaviour ↑ Buildability ↓ Open time	[135]

 \downarrow decreased; \uparrow increased; – same.

3.2. Alternative Binders—A More Sustainable Approach

Despite the use of supplementary cementitious materials (SCMs) like fly ash and slag in concrete production, cement manufacturing remains a significant contributor to global CO_2 emissions. To mitigate this issue, alternative binders such as calcium sulphoaluminate (CSA) cement, limestone calcined clay cement (LC3), and geopolymer cement are discussed in this subsection. For 3DCP, the challenge is the excess surface water that leads to shrinkage, but solutions involving intermediate layers and additives have shown promise. Furthermore, replacing ordinary Portland cement (OPC) with calcined magnesium oxide (MgO) offers sustainability benefits.

The subsection highlights the importance of optimising particle pack density and adding viscosity-modifying agents to enhance printability. Furthermore, it discusses the mechanical properties and durability of cementitious materials, emphasising the potential of geopolymer cement. Despite these advancements, the slow development of 3DCP technologies raises challenges in achieving robust and versatile extrusion systems. Consequently, ongoing research involves a trial-and-error process to balance material properties and advancements in printing technology.

The polemic involving the mitigation of cement production's environmental impact is far from ending. About 800 MT per year of the most common SCMs used in concrete production (e.g., fly ash, silica fume, limestone, and slags) are used, but the cement industry still accounts for 5–8% of global CO₂ emissions [136]. Limitations regarding performance and the scarcity associated with SCMs [48,137] lead to the exploration of globally available materials to formulate new and more sustainable binders to replace OPC, exemplified by CSA cement, LC3, geopolymer cement, and reactive magnesium oxide systems [138].

A significant problem of 3DCP is that excessive surface water expedites shrinkage in the extrusion process, resulting in a weak interlayer bond adhesion. It was found that this deficiency can be overcome by incorporating an intermediate layer of CSA cement, limestone filler, and cellulose fibres that retained excessive water, allowing internal curing, minimising shrinking, and providing mechanical cohesion [132].

Replacing OPC with calcined magnesium oxide (MgO) is an attractive option mainly due to the overall sustainability benefits derived from a greater capacity for CO₂ mineralisation [139] and lower temperatures necessary for clinker production while decreasing the concrete shrinkage strain, upholding good mechanical properties and long-term durability within certain limits [140,141]. Adding suitable additives to printed mortars improves overall performance compared to cast samples [128].

The synergistic effect of calcined clays exemplified by kaolilite, illite, montmorillonite, and limonite (sedimentary rock based on calcite and dolomite) allows the replacement of more than 50% of clinker while retaining mechanical performance and durability [137,142]. The particle pack density can be optimised to improve printability and the increased amount of metakaolin in the calcined clay can overcome problems with extrusion pressure, initial setting time, and compressive strength of the concrete [48]. Furthermore, the addition of suitable amounts of viscosity-modifying agents can reduce the negative impact on the mortar's extrudability of these materials, stabilising the shape of the extruded filaments [143].

The hydration process of OPC results mainly in a calcium silicate hydrate gel (C-S-H) responsible for the time-dependent viscoelastic response to applied deformation [144], impacting the mechanical properties and durability of cement-based materials [145]. On the other hand, geopolymer cement is a binder produced by the alkali-activation of aluminosilicates such as fly ash/rice husk ash, silica fume, clay, metakaolin, GGBS, etc. [146]. Relying on aluminosilicate bonds, this binder is more resistant to acid attacks, with added qualities of high early strength and low shrinkage degrees [147]. Critical reviews of the experimental studies made so far have been recently published regarding the mixture design, printability, and fresh and hardened properties of 3D-printed geopolymers, considering different factors and economic and environmental benefits [148,149]. Further investigation is necessary to eliminate the constraints found [148,150].

All the challenges and constraints related to cementitious mixtures are easily carried over to any conceived material/mixture intended for 3DP, not only because of its particular properties (shape, grain size, density, porosity, water absorption, elasticity, plasticity, hardness, fluidity, mechanical strength, thermal conductivity) but also due to the slow development of 3DCP technologies when it comes to creating robust, autonomous, and versatile extrusion systems that allow the replacement of components to extrude different mixtures. Consequently, all the inherent research and investigation is a trial–error process until a balance is reached [151].

3.3. A Thermal Perspective

Focusing only on the improvement in thermal performance, two strategies can be followed. Strategy 1: In the mixing stage of concrete, a cellular structure like a foam is formed to trap air (low conductivity). The volume fraction of solid material and the void space significantly influence thermal insulation performance [152].

Air-entraining admixtures with surfactant activity (e.g., fatty acids and their salts, alky aryl sulphates or sulphonates, phenol ethoxylates, etc.) contribute to lower the surface tension between water and concrete particles (emulsifying effect), stabilising air bubbles with adequate size (<0.3 mm). The probability of mortar segregation is reduced, the workability is improved, and since water is allowed to expand within those air voids (about 4 to 7% of total volume), the pressure inside the solidified material is relieved, reducing the possibility of shrinkage and crack formation in the concrete surface [153].

The incorporation of a small fraction of oil in mortars, such as recycled engine oil [154,155] or waste cooking oil [156], while delaying setting time and dry shrinkage, reduces water uptake and has a positive impact on air entrainment, reducing pore size, and minimising capillary water absorption and chloride diffusion, hence increasing concrete durability and strength [157] resistance to freezing and thawing, pumpability, and even fire resistance [155,158–160]. Moreover, lipids make the surface of mineral particles hydrophobic, imparting plasticising properties and decreasing the rate of cement hydration [161,162].

Strategy 2: The building envelope that regulates the heat transfer between the indoor and outdoor environments can be enhanced using materials with high thermal resistance, reducing energy costs, and improving thermal comfort [163].

Insulation materials are generally porous, and the thermal capacity is strongly influenced by environmental conditions such as temperature, moisture content, air surface velocity, and other specific properties (e.g., density, ageing time, sample thickness, etc.) [164,165].

Regarding solid waste, several alternatives are available. The challenge in developing mortars with a low environmental impact and good thermal conductivity lies in the necessary balance with proper mechanical and water resistance performances, which generally implies the incorporation of wastes derived from different sources [166].

Based on cost and weight, the most popular materials are natural, synthetic, or organic lightweight aggregates, such as pumice, coal slag, fly ash, and agricultural lignocellulosic wastes [167].

Evidence suggests that paper sludge ash (PSA) can significantly reduce (>80%) the water absorption, sorptivity, and conductivity of concrete when used as a replacement for 12% of OPC or as a hydrophobic surface coating without considerable impact on the hydration, strength, and density of the concrete [157,168].

Pumice is a porous rock of a volcanic origin with very low density. With zeolite, waste is used to produce lightweight asphalt concrete mixtures [169]. It was observed that the mechanical and thermal performances of foamed concrete depend on variations in the density and composition of pumice aggregates. However, reductions of 20% in thermal conductivity can be achieved [170].

It was observed that particular CDW wastes (e.g., concrete, ceramics, sand, glass, wood, etc.) used to replace coarse or fine natural aggregates in concrete mixes showed an

overall thermal performance linearly related to lower density and higher porosity, therefore improving energetic performance for use in buildings [171].

Several reviews about the utilisation of agro-wastes (lignocellulosic materials) are found with the conventional casting process [172–174].

Cement-bonded composites with fibres derived from different sources (e.g., jute, flax, and hemp fibres) exhibited enhanced mechanical performance and thermal and sound insulation [175]. Mortar incorporating date palm fibres showed a decreasing thermal conductivity owing to the increment in the mortar's total porosity [176]. Sand concrete composites with barley straws and wood fibres showed a reduction in thermal diffusivity by 35.5% [172].

Based on the review of cellulose derivatives in printable mortars, crossing cost and thermal performance, incorporating cork and sawdust are promising options [173].

Cork granules embedded in concrete improved lightness and hydrothermal properties [174,177] while establishing the correlation between the cork percentage, hydric transfer properties, and level of cork absorption of water added during concrete mixing [174]. The design of coating mortars with different contents of fly ash (FA), expanded cork granules (ECGs), expanded clay (EC), or a mixture of both as aggregates showed improved thermal performance and water resistance, particularly with FA above 35% and the presence of ECGs [166].

Some restraints need to be overcome by attending to the application of lignocellulosic fibres in cementitious mixtures. The compatibility of these materials with cement is limited, affecting the composite strength and leading to degradation by increased moisture absorption [178]. This effect occurs due to an alkaline attack on lignin, especially the hemicellulose, the compound responsible for natural fibre cohesion, compromising fillers' durability and long-term stability [179,180]. Additionally, the high porosity that develops during mixing plant fibres in the mortar can harm the mechanical properties and performance of the composites [181]. Concrete permeability to liquids is an important property that must be controlled to avoid integrity damage to the structures due to corrosive and dissociative substances [182,183].

Mortars designed with 40% textile fibres to replace sand presented an acceptable mechanical performance, and reduced thermal conductivity and thermal diffusivity, compared to ordinary cement mortar [184]. Elements like coconut coir/husk/shell and corn cob/husk/pith and cotton stalks with high lignin content are suitable for manufacturing insulation panels, and depending on the density of the board, the thermal conductivities can be lower than $0.046~\rm W/(m\cdot K)$ [185]. Similar values were found with materials from diversified sources (e.g., construction waste wool, recycled cellulose (paper or aerogel), recycled PET, recycled wool, wool/polyester, and woven fabric waste) [186]. These materials are all considered high-performance thermal insulators with thermal conductivity values below $0.05~\rm W/(m\cdot K)$, following EN 12664 [187], EN 12667 [188], or ASTM C518 [189] standards.

Many types of plastic waste, like polypropylene, glass fibre [190], textile fibres [191], and metalised plastic fibres [192], have been studied as alternative fillers for thermal insulation building material solutions. Several studies [193,194] have shown that rubber and polystyrene aggregates confer a concrete material elasticity, light weight, vibration damping, durability, and sound and thermal insulation. Lightweight concrete with a 100% recycled plastic aggregate presented thermal conductivity values around 1.1–0.5 W/m·K compared with 1.7 W/m·K of the control concrete [195].

The result of an overview investigating the impact on thermal conductivity of several waste materials (e.g., recycled aggregate, fly ash, rubber, expanded perlite, oil palm shell, palm oil fuel ash, recycled glass, plastic waste, sawmill waste, polyamide, etc.) in concrete composition highlights the relation with the type of materials, degree of replacement, mineralogical characteristics of an aggregate, and surrounding humidity, which influence the water/cement ratio, concrete density, porosity, etc. [196].

A thorough literature review concluded that the type of plastic, the particle size, and the percentage of replacement will condition mechanical strength. Plastics are incapable

of chemical interaction with cement and are hydrophobic by nature. Those characteristics lead to an increased porosity and accumulation of water in the interfacial zone between cement and aggregates, limiting water diffusion inside the concrete, hence inhibiting cement hydration.

3.3.1. Case Studies

Several attempts have been made worldwide to adopt this new technology in civil engineering applications. The first 3D-printed office building was made in Dubai in the United Arab Emirates in January 2016 called "The Office of the Future". The 2000-square-foot office was printed in China, and the 3D-printed cassettes (segments) were shipped and assembled on-site in Dubai. The superstructure was 3D-printed, while the substructure was conventional cast-in-place (CIP) concrete. The concrete mixture consisted of cement, water, glass fibres, and additives, and the printed structure underwent a large deformation, revealing to be necessary the addition of an external transverse post-tensioning to enhance the flexural capacity [197].

Since 2017, several 3D-printed buildings have been reported worldwide. A concrete residence was reported in Moscow, where the main structure took only 24 h to be printed [198]. One of the world's largest and most entirely designed 3D-printed buildings—the Warsan building—was completed in Dubai (640 m², with an entire structure of 9.5 m in height). The structure of the walls was printed using a gypsum-based material, and the fluid was mineral-infused, solidifying in concrete layers [199].

Despite the high implementation and development costs related to the operating scale of 3DP in construction, exemplified by the inherent cost of the necessary machinery, many researchers and entities have boosted the development of this technology through the conception of full-scale architectural components and building elements, considering their thermal behaviour [200].

To enhance thermal performance and improve the mechanical properties of 3D-printed polyurethane (PU) foams, wasted cork from stopper production was added to the composition. For this experience, four types of samples were prepared, each one with different amounts of cork (0 to 5%) and other property values. After thermal conductivity tests, the samples presented λ -values between 0.044 and 0.049 W/(m·K) (for 5 and 0% of cork, respectively). These cork foams have voids in the struts induced by 3DP, which enhances both density and thermal conductivity, a significant benefit since manufacturers and consumers seek to produce lightweight products. However, the production cost of these 3D-printed foams is still unachievable when producing panels for thermal insulation [201].

ETH Zurich, partnered with FenX AG, developed a complex 3D-printable mineral foam formwork with waste. The designed slab is composed of 24 formwork elements with 12 unique shapes. After being printed, they were manually arranged inside a timber formwork and later filled with ultra-high-performance fibre-reinforced concrete (UHPFRC). When the UHPFRC was cured, the timer formwork could be removed, and the final element was completed. The foam elements can be removed and recycled at any time to print the next formwork [202].

A modular 3D-printed vertical concrete green wall system (3D-VtGW) was composed of a double-layered 3D-printed concrete supporting wythe (with a cavity filled with XPS insulation) integrated with a sinusoidal-shaped 3D-printed concrete surface wythe [203]. The space between the two 3D-printed wythes can be filled with soil to support the vegetation (Figure 3). Numerical simulation results suggest that the building with 3D-VtGW revealed a consistent potential for energy saving during summer months and enhanced thermal comfort by reducing wall exterior surface temperature and through-wall heat flux (due to the collective effects of evapotranspiration, heat sink from moist soil, and shield against radiation from vegetation shading).

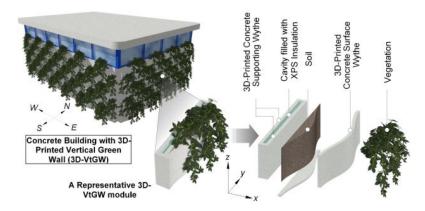


Figure 3. The 3D-printed vertical green wall (3D-VtGW) composition [202].

A study conducted with different scenarios of concrete formulation concluded that introducing agricultural residues, such as hemp in hemp concrete, recycled wood in OPC (IsotexTM), olive cores, or wheat straw in bricks, in general, improves the thermal behaviour and environmental impact in proportion with the concentration [204]. Despite the detrimental effect in the mechanical properties, specifically, due to increased water absorption and biological degradation, scenarios with hemp concrete and Isotex (11.8 and 10.9 kWh/m², respectively) produce an energy consumption analogue to scenarios with conventional concrete containing XPS (10.7 kWh/m²). The same positive effect was found regarding the environmental impact, with hemp concrete exhibiting harmful carbon emissions (hemp plant growth absorbs carbon) and Isotex (0.384 kg of CO_2/kg).

An on-site experiment to assess the thermal properties of a 3D-printed concrete building was conducted by Sun et al. [205] using infrared thermography to evaluate surface temperature distribution and eventual thermal defects (Figure 4). Besides obtaining unsatisfactory results (R-value of 0.31 (m²·K)/W, and effective λ -value of 0.64 W/(m·K) and U-value of 2.27 W/(m²·K)), probably due to the different composition of the 3D-printed mortar in comparison with the conventional one (they usually do not include coarse aggregates), the results showed a markedly non-uniform temperature distribution on the exterior wall surface related to 3DP characteristics (printing path, cross-section design, number of layers, etc.), requiring further work in this field.

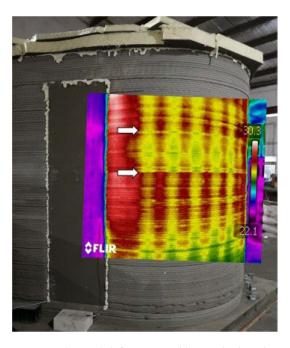


Figure 4. Thermal defects caused by stacked-up layers [204].

3.3.2. Future Challenges

Considering this overview and focusing on the improvement in the thermal performance of 3D-printed building components, some future efforts should be considered [11,206]:

- Some 3D-printed elements have voids in their structure, and therefore, filling them with recycled thermal insulation materials would be a viable option;
- Extruding different materials at the same time, side by side, in a layer-by-layer process, with, for example, a layer of mortar, to provide structural strength and a layer of thermal insulation to improve thermal and acoustic performance ("sandwich wall");
- Changing the composition of the already conceived mortars and incorporating low-conductivity additives to improve its thermal properties while improving other aspects (mechanical, rheological, and printability aspects, for instance).

Additionally, it is relevant to mention that in every theory, potential thermal bridges and high levels of air and water tightness must be investigated and fixed for proper hygrothermal performance and achieving the desired durability of the constructive elements.

As is known, façades display several critical areas responsible for thermal bridges, such as pillars and slabs, connections between a roof façade and ground façade, and corners of exterior walls (windows and door frames, for example). Another essential issue to address is the connection between panels (usually printed separately and then joined on site), usually executed with connectors or male–female fittings; however, these regions should be insulated to prevent heat loss and condensations.

Finally, it will be crucial to consider the adhesion between different materials extruded side by side since their different setting times may comprise the self-supporting role of the wall panels [207]. Using Generative and Parametric Design, different wall solutions can be designed to optimise and enhance the potential of these suggestions [11].

4. Conclusions

Eurostat's in-depth analysis highlights the need for effective waste management practices in the construction industry, as significant amounts of construction and demolition waste (CDW) are generated across Europe and the USA. The circular economy approach to address this challenge involves incorporating waste materials into concrete production, using supplementary cementitious materials (SCMs) such as fly ash, silica fume, and GGBFS. In addition, exploring the potential use of waste materials like clay brick powder, waste glass, and tailings in concrete mixtures expands the range of sustainable materials that can be utilized.

Although SCMs are commonly employed in concrete production, the cement industry is still a major contributor to global CO_2 emissions. Alternative binders such as CSA cement, LC3, and geopolymer cement are being explored to address this challenge. However, adopting 3DCP introduces new challenges, particularly regarding excessive surface water leading to shrinkage. Solutions involving intermediate layers and additives show promise, but further research is needed to optimise printing technology and material properties. "The Office of the Future" case in Dubai exemplifies this approach, where 3D-printed cassettes were assembled using a concrete mixture containing cement, water, glass fibres, and additives, and the Warsan 3D-printed building in Dubai featuring gypsum-based materials infused with minerals to enhance thermal performance.

Future efforts should focus on strategies to enhance the thermal performance of 3D-printed building components. Filling the voids in some 3D-printed elements with recycled thermal insulation materials could be a viable solution. Also, exploring the possibility of extruding different materials simultaneously in a layer-by-layer process, such as a layer of mortar for structural strength and a layer of thermal insulation for improved performance, similar to a "sandwich wall", holds promise. Additionally, altering the composition of mortars by incorporating low-conductivity additives can improve thermal properties while addressing other crucial aspects like mechanical strength and printability. Various waste materials, including lightweight aggregates, recycled plastics, and natural fibres, are

explored for their thermal insulating properties while balancing mechanical performance and water resistance. Nevertheless, addressing potential thermal bridges and ensuring high air and water tightness levels is essential for proper hygrothermal performance and durability. Innovative strategies to optimise thermal performance emerged, such as incorporating wasted cork in 3D-printed PU foams; developing complex mineral foam formwork with waste components, as ETH Zurich and FenX AG demonstrated; and producing modular 3D-printed vertical concrete green wall systems with agricultural residues in concrete formulations. Further studies should address potential thermal bridges and ensure high air and water tightness levels for proper hygrothermal performance and durability.

Looking ahead, a continued exploration of waste utilisation strategies, coupled with advancements in concrete performance enhancement and sustainable construction practices, promises to drive innovation in the construction industry. A more sustainable and resilient built environment can be achieved by harnessing the potential of waste materials and leveraging cutting-edge technologies.

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