ceramics

Review

Sustainable Approaches for the Additive Manufacturing of Ceramic Materials

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Abstract: Additive manufacturing technologies collectively refer to a set of layer-wise deposition methods that typically rely on CAD-CAM approaches for obtaining products with a complex shape/geometry and high precision and reliability. If the additive manufacturing of polymers is relatively easy and scalable due to the low temperatures needed to obtain processable inks, using similar technologies to fabricate ceramic products is indeed more challenging and expensive but, on the other hand, allows for obtaining high-quality results that would not be achievable through conventional methods. Furthermore, the implementation of additive manufacturing allows for the addressing of some important concerns related to the environment and sustainability, including the minimization of resource depletion and waste production/disposal. Specifically, additive manufacturing technologies can provide improvements in energy consumption and production costs, besides obtaining less waste material and less CO_2 emissions, which are all key points in the context of the circular economy. After providing an overview of the additive manufacturing methods which are specifically applied to ceramics, this review presents the sustainability elements of these processing strategies, with a focus on both current and future benefits. The paucity of specific available studies in the literature—which are included and discussed in this review—suggests that the research on additive manufacturing sustainability in the field of ceramic materials is in the preliminary stage and that more relevant work still deserves to be carried out in the future to explore this fascinating field at the boundary among ceramics science/technology, production engineering and waste management.

Keywords: ceramics; 3D printing; waste; circular economy; sustainability

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1. Introduction

Ceramics can be used in various applications due to their multiple properties and are usually formed starting from a mixture of powders using conventional technologies, such as injection moulding or die pressing. However, these techniques may suffer from long processing times, and the machining of ceramic parts is difficult because of their great hardness and brittleness. Other problems that we can find with these materials include the presence of residual pores, which can persist also after sintering, and the difficulty of obtaining a good surface quality and dimensional precision. A solution to the above-mentioned limitations of ceramics processing arose with the use of additive manufacturing, also known as three-dimensional (3D) printing [1]. The implementation of these new manufacturing strategies also allows for the fulfilling of sustainability requirements in terms of materials and technologies, which are becoming increasingly important. In this regard, over the last two decades, there have been significant worries about natural resource depletion and climate change as well as about the resulting ecological repercussions. These burdens have enhanced the appeal of eco-friendly production; in fact, industrial manufacturing processes are responsible for 35–40% of global material consumption and 15% of global energy consumption [2].

The integration of sustainability concepts in the sector of additive manufacturing is feasible and can actually carry important benefits to scientific and technological development, industry, and overall society. This review paper deals with these concepts with a special focus on the processing of ceramic materials. After providing an overview of the different technologies used for the additive manufacturing of ceramics (Section 2), we will discuss how additive manufacturing strategies are environmentally friendly and highly material-efficient (Section 3). Currently available studies on the sustainable additive manufacturing of ceramics will then be presented and discussed (Section 4), before summarizing some concluding remarks and highlights for future research (Section 5).

2. Additive Manufacturing of Ceramics

Additive manufacturing techniques are used to produce components in a point-by-point, line-by-line, or layer-by-layer manner following 3D models, and, thanks to the introduction of this conceptually simple approach, productivity improved and complex objects that were difficult to fabricate could be successfully produced as well. There is an umbrella of technologies under the name of 3D printing that can be divided into three categories: slurry-based, powder-based, and bulk solid-base methods, as shown in Figure 1.

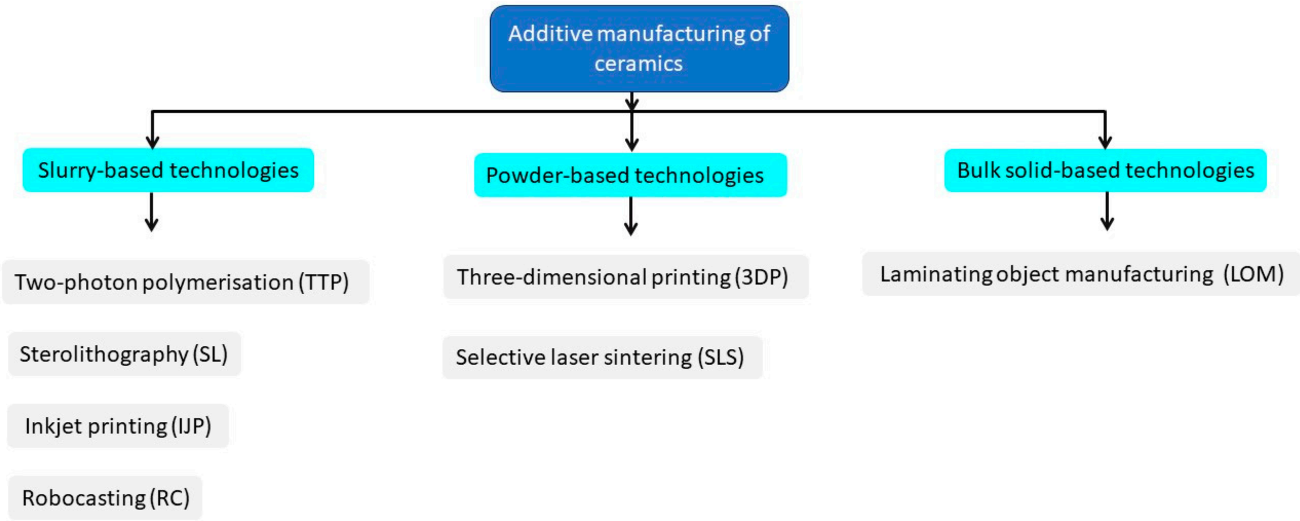

Figure 1. Classification of 3D printing techniques.

2.1. *Slurry-Based Technologies*

Slurry-based ceramic technologies in additive manufacturing mostly involve the dispersion of fine ceramic particles in a liquid or semi-liquid system to form printable inks or pastes. In these technologies, the green body is first printed and then sintered. Pre-ceramic polymers (PCPs) can also be photopolymerised to create polymer-derived ceramic (PDC) components through heat treatment thanks to recent advances in materials science. The ceramic slurry can be 3D-printed by photopolymerisation, inkjet printing, or extrusion [3]. Some examples of these methods include, respectively, stereolithography (SL) and two-photon polymerisation (TPP), ink-based inkjet printing (IJP), and extrusion-based robocasting (RC).

2.1.1. Stereolithography (SL)

In stereolithography (SL), a liquid surface in a vat containing a photopolymerisable monomer and other additives in small quantities is selectively cured by a light source of a desired wavelength and usually proceeds point-to-line, line-to-layer, and then layer-by-layer. The irradiation light is generally in the ultraviolet range [4], but, recently, blue light has also been experimented with, achieving excellent results at the industrial level [5]. The platform supporting the object being produced is lifted or lowered by the thickness of one layer so that polymerisation of one layer is achieved after every irradiation step. A high surface quality is typically acquired by SL; a schematic diagram of this technology is shown in Figure 2a.

In order to make ceramic products, slurries for SL are prepared by adding fine ceramic particles to the photocurable medium along with surfactants and suitable additives so that well-dispersed ceramic suspensions can be obtained. The medium can be aqueous or non-aqueous, the latter being the most commonly chosen type [6]. In fact, because aqueous suspensions would lead to decreased green strength, the majority of suspensions are non-aqueous and resin- or acrylamide-based. Until the full 3D ceramic object is constructed, the ceramic particles are embedded in a cross-linked organic network that is polymerised to create the pre-designed shape of each layer. Once the printing stage has been completed, the green parts need to be sintered at a high temperature to remove the organics and increase the density and the mechanical strength [7]. There is also an indirect version of SL, where resins are used for the investment casting of complex ceramic objects.

The proper design of ceramic suspensions is of primary importance as they should possess an appropriate rheological behaviour as well as long-term stability, because ceramic particles have to be homogenously and effectively dispersed in the medium and the slurry must remain stable over time. As a matter of fact, unstable suspensions with rapid segregation could lead to material inhomogeneity in the printed parts. Another key characteristic of ceramic suspensions is a suitable viscosity for proper flow during the printing process. However, this is challenging because a lower volume of the fraction of ceramic particles is usually required to minimise viscosity and avoid possible segregation, while higher solid loading is preferable for achieving a greater density and less shrinkage [4].

The non-trivial light scattering caused by the ceramic particles added to the suspending fluid is a significant issue for light penetration and also broadens the lateral curing area, which may lead to undesired overlaps [8]. The reflective index of the materials is of particular importance for the curing depth: indeed, this represent a challenge when ceramic particles having greater light absorption and refraction are exposed to the photopolymerisation wavelength. Viscosity and the refractive index ratio govern the final polymerisation rate in the suspensions, which decreases with the increase in these parameters as a result of light scattering and absorption; in this regard, a larger size of ceramic particles yields a greater reduction.

The latest evolution of the SL technology is called digital light processing (DLP) and is based on a dynamic mask to promote the polymerisation of a complete layer of slurry per irradiation cycle (Figure 2b). Digital micromirror devices (DMDs) are used as the dynamic mask generators. A DMD is a chip composed by several thousand microscopic mirrors corresponding to the pixels in the image. DLP carries remarkable advantages such as great feature resolution and higher accuracy and speed of the fabrication process. Complex ceramic structures with extremely fine features may be created using DLP with materials like alumina and bioactive glass, producing relative densities exceeding 90% and mechanical strengths which are superior to those obtained by traditional processing [9].

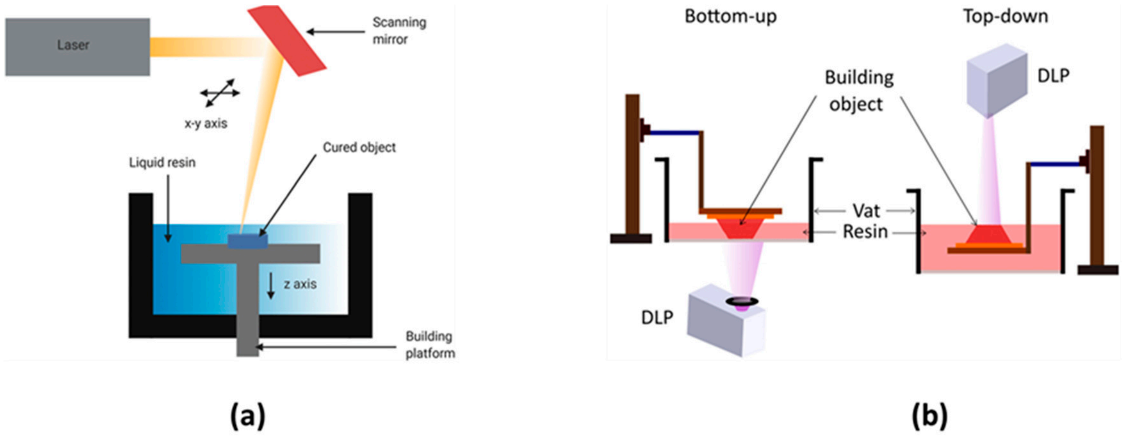

Figure 2. Stereolithographic manufacturing: (**a**) schematic diagram of the SL process (general) and (**b**) schematic diagram of the DLP processes according to a bottom–up or top–down approach. Reproduced from [10,11], respectively, under a CC-by license.

2.1.2. Two-Photon Polymerisation (TPP)

Two-photon polymerisation (TPP), schematically displayed in Figure 3, is used in the nanoscale fabrication of 3D microstructures and relies on the simultaneous absorption of two photons (TPA) of a powerful near-infrared or green laser in a precisely focused point of photosensitive resin [12]. The real benefit of this method is its capacity to use the two photons to polymerise submicrometric focal volumes inside the liquid polymer. Due to the quadratic dependency of the TPA rate on the incoming intensity, it is possible to attain a TPP resolution that is less than 200 nm or close to the diffraction limit. This process was first introduced using polymeric materials but was later also adapted to produce complex 3D microstructures with nanoscale features. In fact, although this technology is limited to employing only polymeric materials, pre-ceramic polymers can be used, too, that are "transparent" and, hence, accessible to the incident laser, since the cross-linking molecules are often utilised at the subsurface sites of the feedstock liquid. On the other hand, TPP does not work with "opaque" ceramic-based slurries, which are frequently used in SL.

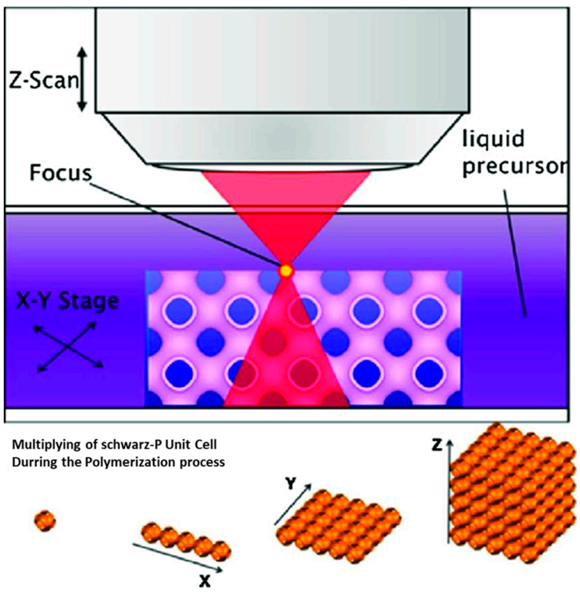

Figure 3. Schematic diagram of the TPP process, showing that the focus of the laser beam guides the polymerisation process. Reproduced from [13] under a CC-by license.

Another drawback of TPP is the long production time due to the extra-fine precision needed for printing very small parts.

2.1.3. Inkjet Printing (IJP)

Inkjet printing (IJP) is a material deposition technique for thin-layer deposition onto substrates: by ejecting liquid-phase materials in droplets from printhead nozzles onto paper, plastic, or other substrates, it produces 2D digital text and images. IJP can be used in two ways: continuous or drop-on-demand (DOD) [14]. The latter is better in additive manufacturing for its higher positioning accuracy and smaller droplet size and can also be attained by depositing ink droplets through thermal excitation or the piezoelectric effect. A schematic representation of IJP is shown in Figure 4.

Due to the extremely small amount of feedstock material utilised, the use of IJP is typically restricted to the printing of miniature pieces. At present, popular commercial printers expel just a few picolitres to nanolitres of ink per droplet. Recently, there has been a transformation of this technique, making it a versatile manufacturing process for the 3D fabrication of multi-layered parts.

The inks are liquid solvents with well-dispersed ceramic particles for direct and selective deposition onto a substrate through the printhead. A point–line–layer–part construction technique is made possible by computer-aided high-precision droplet jetting. After the primary components are properly dried and sintered, a solid ceramic phase can be produced.

The effectiveness of IJP in fabricating ceramics significantly depends on the formulation and physical characteristics of the ceramic powder-containing ink. Specifically, the rheological properties of the ink—including dispersity, stability, viscosity, and surface tension—and the pH value, which must be controlled to avoid the potential corrosion of the jetting system, play a key role. Another characteristic that has enormous importance is the particle size. In fact, nozzle and capillary clogging and obstruction may be avoided by using ceramic particle sizes that are evenly distributed and have a diameter of less than 1/100 of the nozzles [15]. Larger particles should be removed by suitable filters before ink preparation.

Viscosity strongly affects the ejection behaviour of ceramic-based slurries as, if it is too large or too small, it can determine insufficient jetting or too-high velocity, respectively [16]. Ceramic inks frequently have low viscosities due to their low solid loading, resulting in a long drying time and significant shrinkage, which may negatively impact the final accuracy of the printed part. Although increasing the solid loading may be advantageous, doing so may alter the rheological behaviour of the ink, ultimately decreasing printability.

The "printability" of an ink is related to its ability to exhibit a proper printing performance, which should be shown by a high-quality ink compatible with a DOD inkjet printer. A quantification of printability is given by the value of Z [17], which is the reciprocal of the Ohnesorge number Oh (dimensionless):

$$Z = \frac{1}{Oh} = \frac{Re}{We} = \frac{(\gamma\rho a)^{\frac{1}{2}}}{\eta}$$

where $Re = \frac{v\rho a}{\eta}$ is the Reynold number; $We = \frac{v^2\rho a}{\gamma}$ is the Weber number; a is the radius of the nozzle; and ν, η, ρ, and γ are the ejection velocity, the viscosity, the density, and the surface tension of the ink, respectively. The value of Z should be between 1 and 10; if Z is less than 1, viscous dissipation may stop the droplets from ejecting, while, if Z is more than 10, satellite droplets or unwanted tiles could appear.

A huge problem in the IJP of ceramics is the segregation of solid particles from the centre to the edge of a printed structure during its drying. This issue is called the "coffee stain effect" [18] and is caused by convective macroscopic flow into the contact line.

The IJP of the thin functional layers employed as electrodes in energy devices has received a lot of interest recently. Most investigations on this topic focused on ink preparation and layer characterization; from a functional viewpoint, electrochemical results equivalent to those achieved by traditional deposition procedures were obtained by IJP [19].

In summary, IJP is a flexible 3D printing technology that can be used to print tiny ceramic pieces, but it has several limitations when designing complicated structures is a major goal, such as its inability to produce overhanging or hollow structures because of the challenges with support preparation. However, its use in advanced ceramic production has been substantially boosted by the benefits of its low cost, its straightforward processing path, and the multitude of ceramics able to be printed, especially in the microelectronics and energy device field.

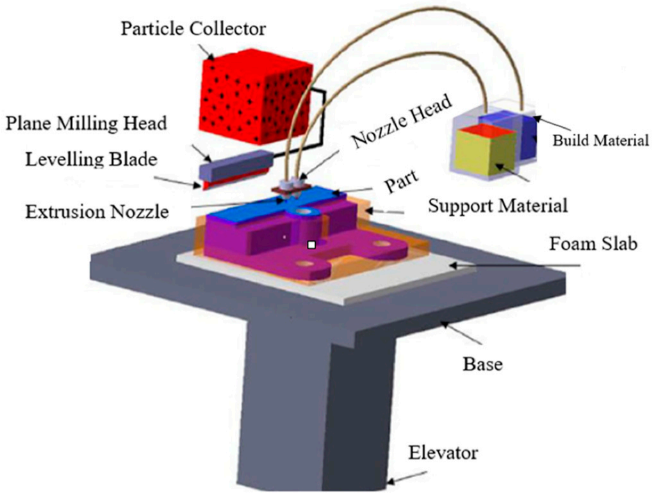

Figure 4. Schematic diagram of the IJP process. Reproduced from [20] with permission.

2.1.4. Robocasting

Robocasting, also known as direct ink writing (DIW), encompasses a set of 3D printing methods that employ extrusion-based ink deposition to create previously designed structures. As shown in Figure 5, robocasting typically utilises a highly viscous paste—similar to the ink used in traditional IJP—at room temperature. A difference between DIW and IJP is the nozzle diameter, being much larger in the former due to the higher viscosity of the materials used. This is a crucial point because the extrusion of the material through the nozzle indeed affects the deposition and quality of the printed filaments. Briefly, the nozzle moves and directly "writes" the desired shape in a layer-by-layer way until the object is built up. The challenges behind ink optimization with the aim of allowing good printability and a satisfactory quality and performance of the final ceramic products have been recently discussed in a comprehensive review paper [21]. Post-printing processes, i.e., debinding and sintering, can be optionally performed depending on whether all-ceramic (with the full removal of organics) or polymer/ceramic composite products need to fabricated.

Robocasting is probably the cheapest and fastest additive manufacturing process among those available. In fact, it does not require the use of special supporting systems for the structures being printed. Freestanding objects with high aspect ratios can be fabricated thanks to the high solid loading and viscoelastic features of semi-liquid pastes [22]. DIW allows for a wide variety of structures to be potentially made, ranging from intricate porous scaffolds to solid monolithic pieces, from micrometre to millimetre scales.

The numerous advantages of DIW have been found to be highly beneficial in the fabrication of bioceramic implants [23]. For example, robocast strong and porous scaffolds promote living-bone ingrowth while supporting host tissue in the body [24,25]. Robocasting also allows for producing reliable models of the body part to be repaired, which effectively reduces the cost, time, and complexity of a surgery.

Pre-ceramic polymers are also suitable for being processed by robocasting [26] and can optimise the rheological behaviour of the feedstock. Furthermore, the introduction of dopants in the ink can help one to tailor the composition, structure, and properties of the final product after heat treatment, as well as the shrinkage.

While robocasting is highly suited for the manufacturing of customised porous ceramics with periodic characteristics and almost no need for surface finishing, the applications of DIW in the processing of thick technical ceramics still is a challenge.

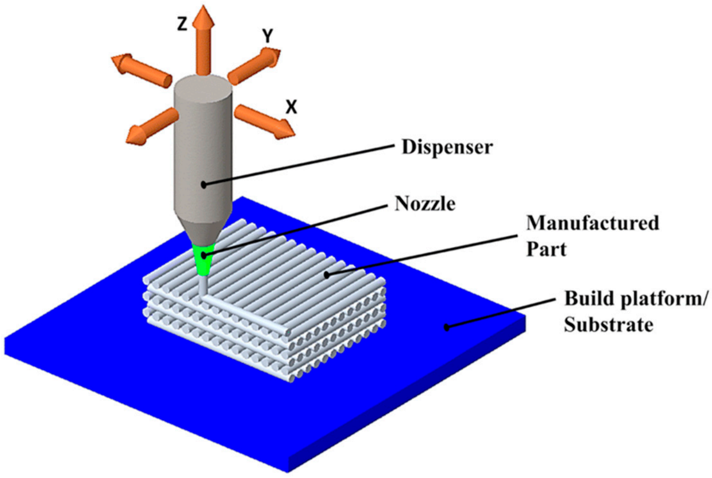

Figure 5. Schematic diagram of the DIW process. Reproduced from [27] with permission.

2.2. Powder-Based Technologies

The majority of powder-based ceramic additive manufacturing technologies rely on powder beds, which frequently use loose ceramic particles as feedstock. The ceramic particles are bonded together either by spreading liquid binders over them or by fusing powder together using thermal energy from a laser beam. There are two types of powder-based 3D processes, i.e., three-dimensional printing (3DP) and selective laser sintering (SLS).

2.2.1. Three-Dimensional Printing (3DP)

An important detail to point out is the difference between the terms “3DP” and “3D printing”. In fact, for historical reasons, the specific “3D printing technology” is still referred to by using the acronym “3DP”, although the expression “3D printing” is now used interchangeably with “additive manufacturing technologies”.

During the 3DP process, a droplet-sized organic binder solution is sprayed onto specific areas of a powder bed using printheads (Figure 6a). Solid layers are then created by solidifying (polymerising) the permeating liquid binder that glues the ceramic particles together. This procedure is repeated until the whole component is built up by supplying fresh powder and spreading it over the previous layer. After that, the loose powder is removed, and the final printed product (green body) is retrieved. Ceramic powders can be deposited in a dry or wet state; in the latter case, before the binder material is applied using an inkjet, the liquid needs to be evaporated. In order to successfully eject the binder solution, certain characteristics have to be fulfilled, such as appropriate rheological properties. The last step is to remove the organic binder by sintering the green, although it may cause the shrinkage of the piece.

The 3DP method was originally designed to rapidly produce components from a larger variety of materials—because no other additive manufacturing techniques could achieve this—and create structural components with quite large sizes, up to several meters [28].

Many studies have been carried out to investigate ceramic powder and binder properties and their interactions and process parameters. The penetration of the binder is controlled by the molecular weight of the organic substance when it covers the ceramic powder bed: specifically, the molecular weight should be less than 15,000 Da to fulfil this function [29]. It was also found [30] that the microstructures, the surface finishing, and the smooth jetting could be improved by optimising the process parameters, such as the surface tension, the viscosity, and other rheological properties of the binder. The resolution of the printed parts also depends on the features of the binder droplet and powdered material. Regarding this last issue, a finer powder is more difficult to spread over the working surface. It has also been observed that lowering the layer thickness reduces the porosity, thereby improving the mechanical properties.

It is not uncommon to find larger pores inside 3DP ceramics with respect to other additive manufacturing technologies. This is a significant problem for the production of high-performance ceramics because they need to be fully dense. Therefore, additional steps in the post-processing stage [31], such as isostatic pressing before sintering, the use of sintering aids, and infiltration of the porous parts, have been proposed to try minimizing such a density limitation.

In summary, the high flexibility of geometrical design without the need of supporting tools for the green bodies being printed is probably the best advantage of 3DP, while the major limitation of this technique applied to ceramic materials is that it can only be used to produce intentionally porous ceramic parts. In general, ceramic products obtained by 3DP have been found to exhibit a lower quality in terms of resolution, surface finishing, density, and mechanical properties compared to those produced via other additive manufacturing techniques (e.g., SL), and, in these cases, additional post-processing steps should be carried out.

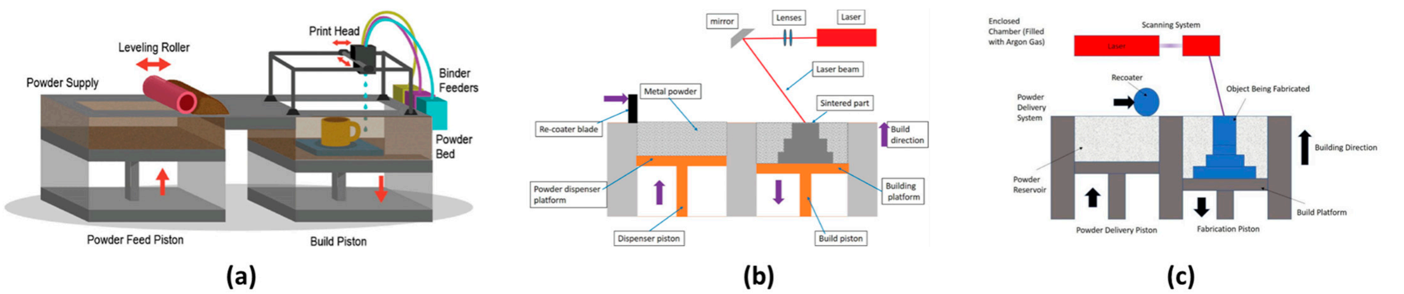

Figure 6. Overview of powder-based technologies: schematic diagrams of (**a**) 3DP process, (**b**) SLS process, and (**c**) SLM process. Reproduced from [32–34], respectively, with permission.

2.2.2. Selective Laser Sintering (SLS)

Selective laser sintering, as shown in Figure 6b, involves the use of a high-power laser beam to selectively irradiate the surface of a target ceramic powder bed, which is heated to allow local sintering to occur. Afterwards, a new layer of ceramic powder is spread onto the previous one, and the process is repeated layer-by-layer. SLS can be performed without the need for additional supporting structures, because the sintered regions are surrounded by the loose powder, which is removed at the end of the whole process. Figure 7 displays a flowchart of the SLS process for ceramics.

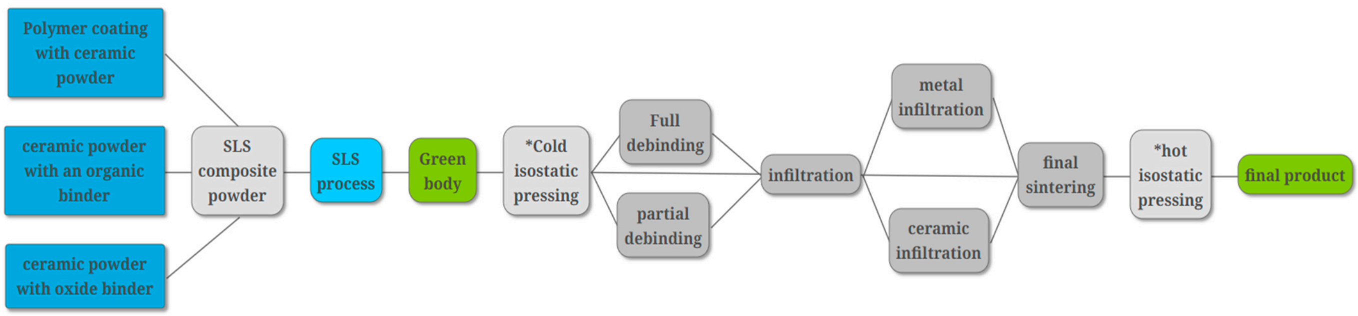

Figure 7. Flowchart for the SLS processing of ceramic components coupled with additional treatments; steps shown with an asterisk (*) are optional.

There are many advantages to using this technology for making ceramics, such as obtaining parts with a reasonable dimensional accuracy and part definition [35]. However, the local densification of ceramic powders is challenging because of the very high melting temperatures, although the high-power laser can potentially generate a temperature which is high enough to trigger the densification process. A variation of the "pure" SLS technique involves coating or combining the ceramic powder with additional substances that have lower melting or softening points and thereby act as binders. This causes the binders to melt and generate a glassy phase surrounding and bonding the ceramic particles together when the laser beam heats the powder bed surface. Organic binders can be used, too, which are removed by high-temperature firing in a furnace once the SLS process is completed, thus allowing for the production of all-ceramic products. However, firing cannot remove the inorganic binders (e.g., glass), which will remain in the final product and may interact with the matrix powder to create a secondary phase. Indeed, their presence has to be taken into account in terms of material performance (e.g., they can cause a slight decrease in the mechanical properties).

The two major disadvantages of SLS are the high shrinkage and high porosity remaining in the final components [36]. However, structural ceramics should be made almost fully dense to perform at their best from a mechanical viewpoint. In this regard, combining SLS with infiltration/isostatic pressing (as also performed for 3DP) can increase the density of the final ceramic parts [37].

It is worth pointing out that the properties of the parts produced by SLS are affected by a number of factors associated mainly with the feedstock materials and laser–material interactions. Good flow characteristics are required for the matrix and binder powders, which are better for particles of a spherical shape and a micrometric size [38].

With regard to the reaction between the laser beam and the material, there are various dynamic conditions appearing at the level of localised microscopic interaction volumes that need to be considered during the rapid laser-fusing process. This process ultimately influences the microstructure, geometry, and mechanical properties of the final parts. A key factor is the energy of the laser applied to the ceramic powder bed during SLS, which is directly associated with the laser power and the scanning speed. The laser energy to be applied depends on the powder composition (ceramic + binder), the thermal properties of the powder (e.g., melting point and thermal conductivity), and the packing porosity of the powder bed. A too-low laser energy yields insufficient fusion of the binder with poor bonding of the adjacent layers, resulting in a low strength of the green body. On the contrary, a too-high laser energy leads to excessive melting and evaporation of the binder (if it is organic), thus resulting in geometrical and/or functional damage of the part [39].

A variation of SLS is selective laser melting (SLM), schematised in Figure 6c. It works similarly to SLS, but it employs laser sources with significantly higher energy densities; therefore, ceramic powders can be fully melted without the need for low-melting binders, and fully dense ceramics can be obtained. The SLM of ceramics, however, is much more difficult compared to other materials. In fact, it is very challenging to produce a non-porous, isotropic ceramic body starting from porous microstructures with rough surface finishes and low dimensional accuracy.

2.3. Bulk Solid-Based Technologies

The most common technique among the bulk solid-based technologies is laminated object manufacturing (LOM), which is schematised in Figure 8. The process typically entails layer-wise bonding of one cut sheet on top of another, which has been pre-coated with adhesive agents, to create 3D components, followed by computer-controlled laser cutting of thin sheets of the materials into cross-sections in accordance with sliced digital CAD models. Real-time heating and mechanical compression can be used to bond and laminate neighbouring layers. The procedure can be completely automated thanks to the continuous rolling of long green ceramic sheets onto the working platform. Before each item is laminated, excess material surrounding it is removed layer-by-layer.

Finally, high-density components can be produced following additional binder removal and high-temperature sintering.

The main advantage of LOM is the elimination of deformation and distortion thanks to the low thermal stress in the manufacturing process [40]. However, the main weaknesses of this technique are associated with the weak interfacial bonding between layers, which may lead to delamination, interfacial porosities, and anisotropic properties along the planar and building direction [31]. Another disadvantage of this process is its restricted use, as only laminated sheets can be used. A further drawback is the poor surface quality obtained in the parts, especially when dealing with round-shaped surfaces; however, it might be overcome by combining LOM with hight-speed cutting techniques [41].

Owing to all these shortcomings, there has been a limited technological progress in laminated object manufacturing. Specifically, the main obstacles are the complex geometry of advanced ceramic components and their miniaturisation below the micrometric scale, because the ceramic LOM process typically produces parts of large dimensions with limited structural complexity.

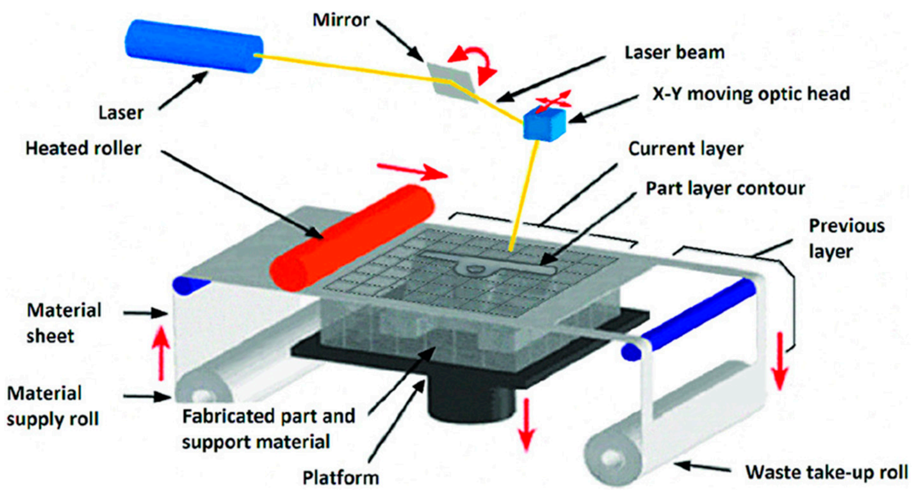

Figure 8. Schematic diagram of the LOM process. Reproduced from [42] under a CC-by license.

2.4. Other Additive Manufacturing Approaches

The technologies described in the previous sections allow one to obtain final products that are entirely made of ceramic. Composite materials comprising ceramic inclusions embedded in a soft polymeric matrix can be fabricated using fused deposition modelling (FDM), which is also known as fused filament fabrication. FDM is a 3D printing technology that uses thermoplastic filaments to create solid objects layer-by-layer, as schematised in Figure 9. The equipment for this process is akin to the process of a 3D printer extruding molten material through a nozzle to build up the final shape: each layer cools and solidifies, fusing with the previous layer [43]. Indeed, FDM cannot be applied to directly process ceramics due to the ultra-high melting temperatures for such materials. Common FDM materials include polymers such as poly(lactic acid) (PLA), acrylonitrile butadiene styrene (ABS), polyethylene terephthalate glycol-modified material (PETG), etc. In recent years, FDM has been applied to produce hydroxyapatite- or bioactive glass-containing composite parts with intricate geometries for biomedical applications, such as porous scaffolds for bone tissue engineering [44,45]. One study also reported the printing of Si_3N_4 ceramics by extruding a feedstock composed of ceramic powder and organic binder through a nozzle and depositing it layer-by-layer [46]. FDM for ceramics opens up exciting possibilities, bridging the gap between traditional ceramics and modern additive manufacturing. As technology advances, we can expect even more breakthroughs in this field.

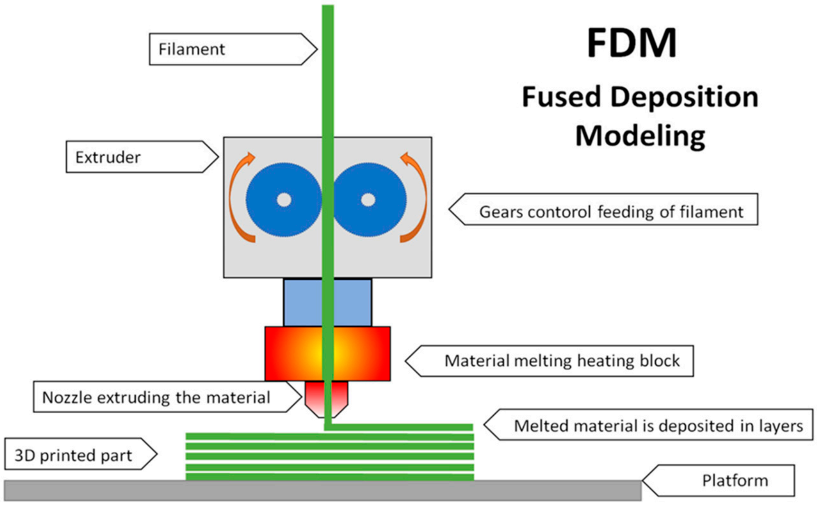

Figure 9. Schematic diagram of the FDM process. Reproduced from [43] under a CC-by license.

3. Sustainability of Additive Manufacturing

In general, additive manufacturing technologies are environmentally friendly [47] and highly material-efficient because of waste material minimization. Specifically, additive manufacturing techniques minimise the life cycle material mass and energy spent in traditional subtractive processes by reducing the scraps. There are three main sustainability advantages of additive manufacturing compared to conventional processes [48], i.e., (i) the ability to fabricate products with a more efficient operational performance, (ii) the need for lower amounts of raw materials throughout the supply chain, and (iii) the decrease in the use of ineffective and energy-intensive production methods.

3.1. Sustainability Pillars for Additive Manufacturing

The sustainability pillars of additive manufacturing are the environment, society, and the economy, and each of them is related to some specific aspects of this process [49]. The environmental pillar is related to resource consumption, pollution control, and waste management; the economical pillar concerns the evolution in the market, supply chain management, production, direct and indirect costs, and productivity; and the social and personal health pillar focuses on social benefits, labour market development, product quality, consumers' acceptance, healthcare improvements, ethical development, intellectual property issues, and patents.

This analysis is essential because adopting additive manufacturing technologies would deeply alter the production process. In fact, there could be many aspects to be considered when using these methods, such as more efficient production systems, the integration of new materials, the assumption of new business models, and the enforcement of new manufacturing processes. Using additive manufacturing could result in important savings throughout the production and usage stages due to lower energy usage, material inputs, and handling and shorter supply networks.

3.1.1. Environmental Pillar

The additive manufacturing sector uses resources primarily through exploiting materials and energy. When compared with traditional production processes, additive manufacturing technologies are more efficient with regard to materials' consumption. On the other hand, low productivity and the other accessory needs of various additive manufacturing techniques may result in increased energy usage [50,51]. The environment is kept clean, and production is moving towards being more ecologically friendly by using additive manufacturing technologies because cutting fluids, lubricants, and other materials are not required. Overall, the environmental effect of additive manufacturing has been mostly investigated in three areas: resource use and consumption, waste management, and pollution control.

Resource consumption might be divided in material and energy use. Material utilization includes basic (e.g., ceramic powder) and secondary materials (e.g., supporting structures), protective gas, and cooling water, which is significantly less than that used in conventional manufacturing. It has been estimated that additive manufacturing can be 97% material-efficient [52]; however, in real scenarios, this value is considerably lower.

Traditional manufacturing is subtractive, which means that a lot of waste (e.g., ceramic dust) is produced before completing the fabrication process, while additive manufacturing may reduce this waste by 90% [49]. However, not much action has been taken to evaluate this waste material; little actions have been taken to look into waste management, despite it being at the forefront of promoting sustainable manufacturing. This may be due to the current still-modest share of the additive manufacturing industry. There is little evidence to show how much waste the additive manufacturing of ceramics generates overall [53].

In terms of pollution, waste might be in form of gas, liquid, or solid. The generation of particles during printing and the risks associated with powder handling and use deserve to be studied for additive manufacturing processes, as carried out for conventional processes [54]. Compared to subtractive manufacturing, additive manufacturing uses fewer potentially dangerous chemicals, such as forging lubricants and cutting fluids.

In summary, additive manufacturing has (at least) six key environmental advantages [55]: (i) less raw resources required throughout the supply chain, (ii) lower energy consumption, (iii) lower waste production, (iv) a reduction in the weight of the goods used in transportation, (v) enhanced operational performance, and (vi) the decentralization of production (closer to the point of consumption).

3.1.2. Economical Pillar

There are two major factors that may affect economic performance when using additive manufacturing techniques [56]: energy use as a direct cost and investment profitability. Compared to traditional fabrication methods, the additive manufacturing of ceramics reduces the costs in new product development and volume manufacturing, but—at least so far—it cannot compete for mass production and is not suitable for bigger batch production systems. This is because of the concept of the economy of scale, where production and fixed cost per unit have an inverse relationship. In other words, the fixed cost per unit decreases when manufacturing volume increases, since the costs are spread out over a larger number of items. Because mass-manufacturing has lower unit costs, additive manufacturing is less competitive as production volumes rise [56]. This suggests that additive manufacturing is more convenient than traditional manufacturing for low-volume production, although it depends on a machine's capacity, the material's quality, and the quality requirements needed for the final product. Additive manufacturing techniques also do not require tools or moulds, which may lower the costs and be an advantage if we wanted to produce a single customised or complicated design.

An economic disadvantage of additive manufacturing technologies for ceramics is that some the machines required to process such materials are typically expensive (e.g., SL and SLS), although this limitation is expected to decline as these techniques become more popular [57].

By replacing several elements made up of various materials with a single integrated assembly using additive techniques, assembly-related problems with respect to the cost, time, and quality can be minimised or completely eliminated. Redesigning a product to achieve the best strength-to-weight ratio is also possible, while still satisfying the functional needs and using less material. Furthermore, when the component design is ready, the manufacturing process may start right away: the cost reductions accompanying these technologies arise by reducing the timeframe between design and production.

The business models for additive manufacturing technologies concentrate on the ways in which businesses generate value, for whom they provide it, and how they capture it [58].

Additive manufacturing also opens up new business opportunities in high-tech sectors, along with the resultant extensions in a product's life cycle, thus encouraging businesses to embrace product–service business models. Product-oriented company models may incorporate a range of product-related services such as the distribution of consumables and spare parts, updates/upgrades, remanufacturing, recycling, etc. [59]. The relative paucity of skilled machine operators, however, is now impeding the viability of providing these kinds of services affordably.

Briefly, additive manufacturing technologies have (at least) six "sustainable advantages" from an economic perspective, which can be summarised as follows [57]: (i) smaller production runs of bespoke goods are more economical compared to traditional mass-manufacturing techniques; (ii) tools and moulds are not required for direct manufacturing from 3D CAD models (no associated relevant costs); (iii) digital data for designs may be readily exchanged and modified, reducing the cost for altering the components; (iv) additive manufacturing technologies reduce waste by recycling leftover materials (e.g., ceramic powder and resin) from the production process; (v) easiness in designing innovative, complex structures like lattices, foam-like structures, etc.; and (vi) decrease in inventory risk due to the absence of unsold finished products and increase in revenue flow due to payments made prior to production.

3.1.3. Social and Personal Health Pillar

The societal impact of additive manufacturing on social sustainability is poorly understood, with studies focusing mainly on worker's health and working conditions [54]. Additive manufacturing may indeed be safer than traditional methods, but toxicity-related effects still need to be comprehensively investigated [51]. Direct digital manufacturing technologies enable the democratization of production, transforming individuals from passive consumers to prosumers in the global industrial society.

Traditional manufacturing processes generate, also for ceramics, a range of air/water pollutants, noise, fluid spills, and waste powders, all of which pose serious risks to workers' health and the environment [59]. Additive manufacturing techniques could minimise or even stop these problems; however, they might lead to new health problems. For instance, current research on organic chemical emissions from 3D printing (e.g., the use of solvent, photocurable resins for SL, etc.) suggests that they may have detrimental effects on the cardiovascular and pulmonary systems. With regard to the effects of ceramic powder pollutants, although particle exposure may be hazardous to human health, their concentration is dependent on several parameters such as size (micro- or nano-range), ventilation, room size, and the features of the printer cabinet [60].

As a result of all these problems, the possible toxicity, environmental dangers, and chemical degradability of the solvents used during printing processes still remain matters of concern.

3.2. Sustainable and Circular Economy Approaches for Additive Manufacturing

Materials' selection and sourcing are crucial stages in creating a more informed and sustainable supply process. A circular economy approach, based on the six key words "reduce, reuse, recycle, redesign, recover and remanufacture", aims to maximise resource use while reducing waste generation and disposal [61]. This approach is an alternative to the conventional linear economy paradigm and focuses on sourcing resources like recycled, biodegradable, and renewable materials suitable for additive manufacturing systems [62].

The circular economy concept and additive manufacturing technologies may be combined together, with the aim of adding value to a product by carefully considering the sourcing and selection of the materials. The relationships between the three sustainability pillars described in the previous sections about additive manufacturing and their integration in the circular economy approach are schematically depicted in Figure 10. In general, this valuable integration can promote scientific and technological innovation, provide a smart way to reduce waste production and emissions, and enhance product performance and design. Therefore, it should be imperative for governments and companies to take the initiative of promoting sustainable practices [59].

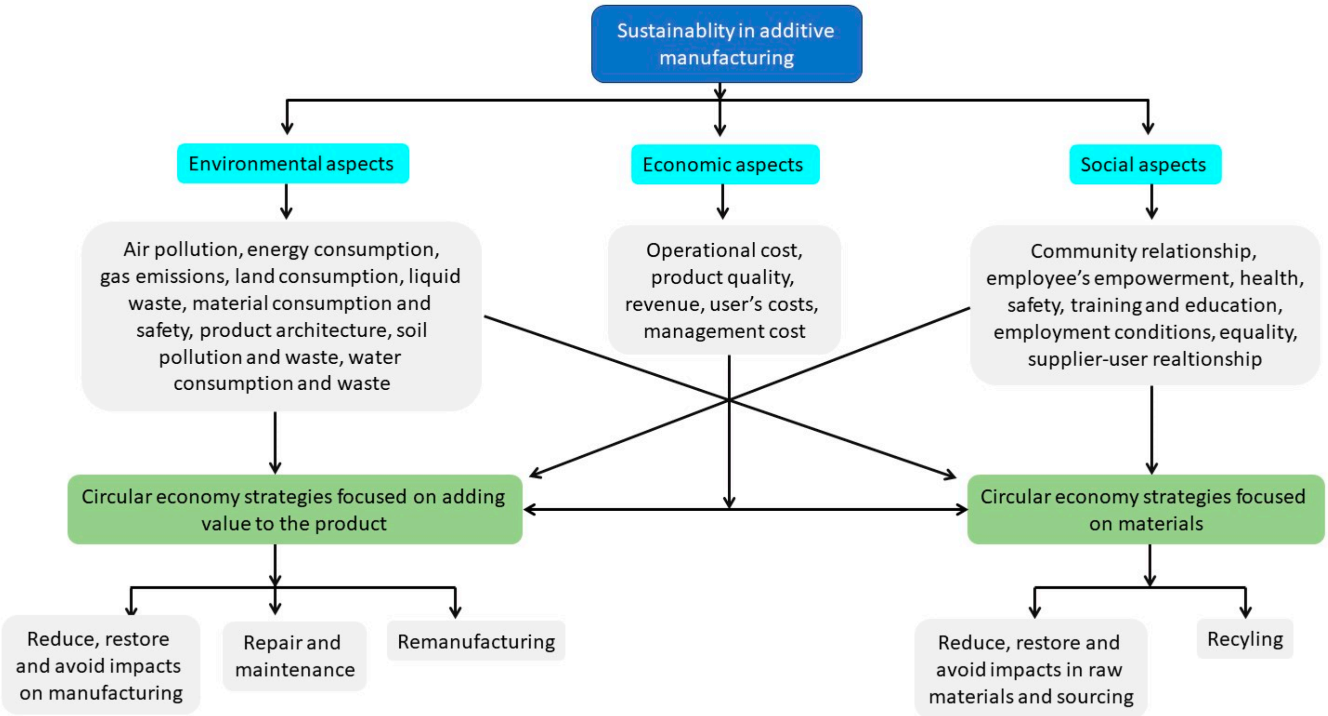

Figure 10. Integration of additive manufacturing and the circular economy concept from environmental, economic, and societal viewpoints.

4. Possible Recycling Options and Sustainable Materials in the Additive Manufacturing of Ceramics

There is a paucity of specific studies in the literature regarding the incorporation of circular economy/sustainability concepts in the additive manufacturing of ceramic materials, which suggests that this field is at its beginning stage. From a general viewpoint, most studies deal with waste or recycled glass. Marchelli et al. [63] provided some general guidelines on how to adapt additive manufacturing technologies to process both virgin and recycled glass. Klein et al. [64,65] described the formulation of a new material to be used in a 3D printing extrusion process to create optically transparent glass-based products.

Some of the available studies also deal with composite materials, in which a glass phase is embedded in a polymeric or cementitious matrix; it is worth noting that most of these works are aimed at construction/building applications. In this regard, Andrew et al. [66] and Ting et al. [67] proposed the use of recycled glass as an aggregate for the 3D printing of concrete.

A study by Cueva et al. in 2022 [68] was aimed at developing 3D-printed lightweight concrete using waste glass as a replacement for natural aggregates, along with expanded thermoplastic microspheres, in an effort to enhance concrete's properties for construction. The modifications in the mix's composition to maintain flowability during printing and the subsequent impact on thermal conductivity and compressive strength were illustrated and discussed, showing a path toward optimizing this material's properties for construction applications by implementing sustainable additive manufacturing strategies.

A recent study reported in 2023, focusing on waste glass powder as a partial replacement for cement in 3D-printed concrete, provided valuable insights into the trade-offs between material properties and sustainability goals. The correlation established between fluidity and rheology for assessing printability also offered a practical approach toward sustainable construction practices [69].

A study about the performance of innovative cement–glass composite bricks incorporating recycled poly(ethylene terephthalate glycol) and a 3D-printed internal structure also revealed the functional advantages that can be obtained by the implementation of sustainable materials, leading to final products with enhanced thermal insulation and mechanical properties [70].

Indeed, a balance between recycled contents and mechanical performance can be more or less difficult to achieve depending on the materials and technologies used, as suggested in a study about the optimization of extrusion 3D printing using waste glass fibre-reinforced polypropylene composites [71]. This evaluation indicated the significance of finding optimal content ratios for industrial thermoplastic composites, supporting the role of extrusion 3D printing in circular economy initiatives.

Waste glass was also proposed in contexts other than construction and building. A study focusing on recycling end-of-life glass fibre-reinforced wind turbine blades for composite materials processed by 3D printing highlighted a strategic approach to reuse waste in the composite industry. The analysis of the specific stiffness, tensile strength, and failure strain resulting from the incorporation of various fibre contents contributed to the better understanding of the mechanical behaviour of these materials, showcasing the potential for improved structural stiffness through recycled fibres [72].

A couple of studies dealing with non-glassy materials were also reported in the literature. For example, the use of calcite-based mussel shells in additive manufacturing processes represents an ingenious effort to adapt waste materials into functional 3D-printable components. The researchers optimised the composition of the printable inks through dosing mussel shell powder and sugar water and evaluated key aspects of the printed parts like their strength, density, and recyclability [73].

The anisotropic behaviour in geopolymer mortars made from construction and demolition waste using additive manufacturing was reported in 2022 in another study, which highlighted the complexities involved in material performance due to the layer-by-layer printing process. Understanding the influence of the different waste materials and alkaline activators on the mechanical properties of printed products can indeed contribute to potential advancements in optimizing structure performance [74].

5. Conclusions and Outlook

Additive manufacturing technologies carry an enormous potential for improving the performance and quality of both high-tech and popular products and can also fit well with the sustainability requirements that are becoming imperative for coping with the needs of a world that has finite resources but infinite desires. In fact, improvements in resource efficiency, more effective manufacturing processes, the use of new materials, new manufacturing techniques, and new business models are all made possible by making use of additive manufacturing's capabilities. Furthermore, implementing additive manufacturing approaches allows one to save money during both the fabrication process and the use of the final product. Less handling and material inputs are used during the production process, which results in cost reductions along with shorter supply networks. This further highlights the important contribution of sustainable additive manufacturing towards addressing global environmental concerns.

On the other hand, there are significant challenges with sustainable additive manufacturing, and, as a result, suggestions for process adjustments as well as new materials have to be used. These challenges are particularly significant for ceramic products, which typically require a post-printing stage of thermal consolidation (sintering) with its associated energy consumption. Possible strategies for trying to follow a circular economy philosophy include waste incorporation as well as the use of local ceramic material in order to reduce all the consumption related to transportation. As an alternative to the traditional linear economy paradigm, the circular economy strategy focuses on finding resources such as recycled, biodegradable, and renewable materials also suitable for additive manufacturing processes. This approach represents a system in which the resource loops are closed.

The studies published so far about the sustainable additive manufacturing of ceramics are rather limited and mainly focus on reusing local and/or waste materials. Studies utilizing powdered mussel shells, broken wind turbine blades, waste glass, and construction debris reveal innovative approaches to additive manufacturing. These investigations rigorously analyse key properties like strength, density, and further recyclability potentials and highlight the pivotal link between sustainability goals and functional performance metrics, thus guiding the optimization of materials for structural purposes, thermal insulation, and mechanical robustness in the printed components.

However, other important aspects, such as a more systematic development of sustainable printable ink formulations—e.g., by means of the careful selection of sustainable polymers and/or the replacement of organic solvents with aqueous media—still need to be investigated. In this regard, it cannot be ignored that some additive manufacturing technologies may increase the proportion of organic materials in the raw materials when, for example, polymer-based slurries or inks need to be used (e.g., in stereolithographic methods and robocasting). This leads to an increment in carbon dioxide emissions during post-printing thermal treatments (debinding and sintering), which deserves to be taken into account and quantitatively evaluated, perhaps by using appropriate life cycle assessment (LCA) methodologies.

This gap highlights a great opportunity for future research and development in expanding sustainable practices for the making of ceramics. These perspectives herald a new era, emphasizing the fusion of innovation with sustainability in production processes and paving the way towards eco-conscious additive manufacturing.

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