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Biochar

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Biochar–polymer composites for 3D printing: a review

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Abstract

Biochar, a bio-based co-product of biofuel production via thermochemical conversion, holds potential as a filler for polymer composites to reduce costs, improve thermomechanical properties, and aid in environmental remediation. 3D-printed biochar composites have received growing interest over the past few years but have experienced difficulties such as poor layer adhesion and nozzle clogging. Currently, no literature review examines 3D-printed biochar composites and related biochar properties in-depth. This work summarizes and discusses recent studies on 3D-printed polymer and biochar composites and examines their mechanical, thermal, and additional properties that result from each study. Technical challenges in printability, such as nozzle clogging from particle size and biochar aggregation, are also discussed. Furthermore, this work discusses the variability of biochar properties resulting from the pyrolysis conditions and feedstock choice in relation to potential 3D printing outcomes. In particular, several studies reported that high lignin feedstocks could be candidates for 3D printing. The post-processing approaches of the biochar via physical and chemical methods are also introduced. Ball milling appears to hold the most promise for physical treatments due to its tunability of particle size, surface area, and functional groups, while chemical treatments with acids or alkalis are used to tailor biochar porosity and wettability. Overall, it was determined that future research needs to be done relating biochar production and post-processing methods to resulting 3D printing parameters as the number of studies is limited.

Highlights

- Biochar can be used as a renewable polymer filler in 3D printing; however, it has a few challenges.
- These challenges include nozzle clogging, die swelling, and layer adhesion.
- Biochar can be tailored to improve printing through preparation conditions and post-processing techniques.

Keywords Fused deposition modeling, Additive manufacturing, Pyrolysis, Biocarbon, Renewable, Filler

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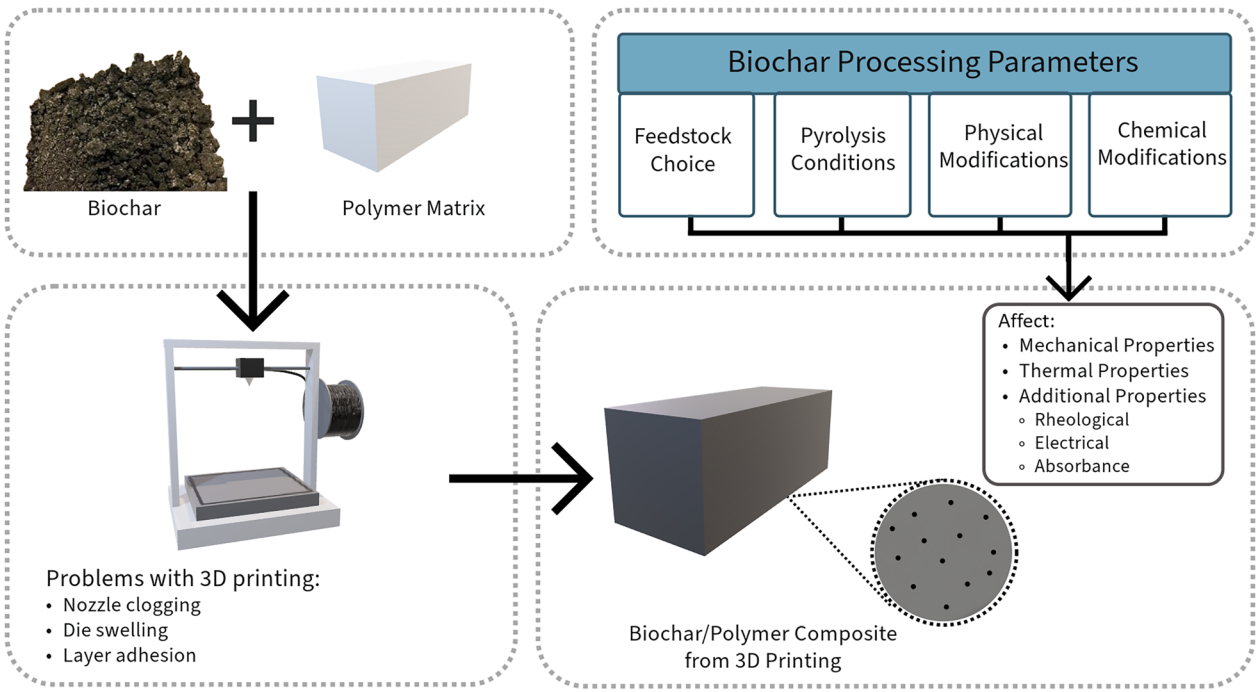
Graphical Abstract



1 Introduction

Biochar (BC) is a carbonaceous material obtained from various resources, such as lignocellulose (Ayten and Oskay 2022; Bamdad et al. 2019; Baronti et al. 2014; Choudhury and Lansing 2020; Feng et al. 2020; Inyang et al. 2014; Ma et al. 2021; Olu-Owolabi et al. 2021; Sahota et al. 2018; Shimabuku et al. 2016; Wang et al. 2021; Xiong et al. 2017; Yazdani et al. 2019; Zhao et al. 2022; Zhong et al. 2019; Zhou et al. 2018), organic waste (Son et al. 2018; Yu et al. 2018), and livestock manure (Glazunova et al. 2018; Nan et al. 2021). It can be formed from these feedstocks via thermochemical (e.g., pyrolysis, gasification, and hydrothermal carbonization) processing methods. In many conventional biofuel production processes, biochar is not a primary product; however, it has several promising properties, such as high porosity, high specific surface area, charged surface, long-term stability, and cation exchange capacity that make it a valuable material (Ding et al. 2016; Zhao et al. 2020). Biochar has been widely researched as an absorbent for dyes (Inyang et al. 2014), medication (Olu-Owolabi et al. 2021; Shimabuku et al. 2016), heavy metals (Son et al. 2018), and gases (Bamdad et al. 2019; Choudhury and Lansing 2020; Sahota et al. 2018; Zhou et al. 2018). Furthermore, it has been explored for electrocatalytic applications (Ayten and Oskay 2022; Ma et al. 2021; Zhao et al. 2022; Zhong et al. 2019) as well as soil remediation (Baronti et al. 2014; Glazunova et al. 2018).

The inclusion of biochar in polymer composite applications has recently been highlighted as an alternative to traditional carbonaceous fillers. Table 1 lists recent research being done on polymer–biochar composites in fields such as packaging, aerospace, automotive, construction, energy, and defense. These applications can be manufactured under multiple processing routes, including injection molding (Das et al. 2016; Zhu et al. 2020), melt blending (Das et al. 2016; Tolvanen et al. 2019; Zhang et al. 2019; Zhu et al. 2020), hand lay-up (Matykiewicz 2020), thermal phase inversion (Ghaffar et al. 2018), and electrospinning (Taheran et al. 2017). In particular, biochar-based polymer composites have gained increased attention due to their utilization of pyrolyzed biomass materials, as they make composite materials more bio-based, sustainable, and environmentally friendly while utilizing a byproduct. Additionally, the inclusion of biochar into the polymer matrix can improve the mechanical properties and thermal stability as well as add desired functionalities (e.g., electrical conductivity). Biochar is less costly than traditional carbonaceous fillers and has the potential for high compatibility with many polymer matrices (Bartoli et al. 2022). Since biochar is a lightweight and low-density material, it is promising for

Table 1 Summary of biochar applications in polymer composites

Polymer	Biochar source	Processing method	Application	Reference
Polypropylene (PP)	Landfill pine wood waste	Melt blending, Injection molding	Food packaging, Interiors of airplanes and automobiles, Fire retardant materials	Das et al. (2016)
High density polyethylene (HDPE)	Poplar wood	Melt blending, Hot pressing	Packaging, Automotive sectors	Zhang et al. (2019)
Polyamide 6 (PA 6)	Bamboo	Melt blending, Injection molding	Furniture, Construction, Packaging, Automobiles, Aerospace, Bridges	Zhu et al. (2020)
Agave sisalana fiber-reinforced epoxy composite	Coffee waste stream	Compression molding, Thermo-mechanical cure	Automotive, Civil construction, Naval sectors	Zuccarello et al. (2021)
Carbon fiber-reinforced epoxy composite	Fish scales	Compression molding	Construction, Automotive, Aerospace, Defense, Wind energy	Rajendran et al. (2025)
Polylactic acid (PLA)	Bamboo, Bamboo cellulose nanowhiskers	Solution casting	Agricultural sun shading films, Package films	Sheng et al. (2019)
Graphite-polylactic acid (PLA) composites	Pine	Melt blending, Hot pressing	Wearable and portable devices (electromagnetic interference shielding materials)	Tolvanen et al. (2019)
Polyvinylidene fluoride (PVdF)	Wood biomass	Thermal phase inversion	Membrane (Pollutant management)	Ghaffar et al. (2018)
Polyacrylonitrile (PAN)	Pine white wood	Electrospinning	Membrane (Wastewater treatment)	Taheran et al. (2017)

weight reduction in the vehicle or aerospace fields. Its pyrolysis treatment provides improved thermal stability, where biochar composites have been investigated for fire retardant properties (Das et al. 2017). Furthermore, its porosity has allowed it to be explored as an absorbent membrane with polyvinylidene fluoride for dyes such as Rhodamine B (Ghaffar et al. 2018). Additionally, its bio-based derivation and environmental sustainability have influenced its use in construction material composites (Rajendran et al. 2025). Overall, biochar can be tailored in its properties by selecting different feedstock sources and production methods, translating into varying properties for polymer composites.

In addition to these polymer–biochar composite processing techniques, the study on the use of biochar in polymer matrices through 3D printing has attracted more attention. This additive manufacturing technique provides a unique concept in polymer product manufacturing, in which particular shapes can be computationally designed and printed using a composite filament. In terms of popularity, fused deposition modeling (FDM) has been highly researched due to its user accessibility as well as its wide range of part production options and designs (Komal et al. 2021). Researchers have been able to compare 3D printing technology with traditional manufacturing methods to examine composite property changes. Primarily, polymer thermal properties were found to improve due to the short exposure time to high temperatures during processing, in comparison to injection molding (Komal et al. 2021). In some cases, the polymer remained thermally stable, though it did not improve significantly compared to traditional methods (Askanian et al. 2018; Cisneros-López et al. 2020). Only a few review articles have focused on biochar/polymer composites but without discussing 3D-printed biochar/polymer composites as the main focus of their article (Hassan et al. 2024b; Li et al. 2023b). Hassan et al. investigated the recycling of polymers and their 3D-printed properties, including biochar composites for sustainable polymer fillers (Hassan et al. 2024b). Li et al. discussed the use of biochar in the printing category as a whole, including dye absorbent, 3D printing, and pigments (Li et al. 2023b). However, a review by Bolanakis et al. briefly examines 3D printing of biochar composites in regards to polymer types, though the authors do not specify correlations in biochar production and post-processing to composite properties (Bolanakis et al. 2024).

Despite the great potential of biochar-based polymer composites, they have some challenges for practical applications. Firstly, since biochar can be produced from a wide range of feedstocks, the feedstock variability makes its industrial applications challenging. Secondly, the application of biochar in composites is still relatively new, so the correlations between biochar and composite properties are not fully understood yet, especially in 3D-printed composites. With 3D printing, there is also the drawback of ensuring good printability of the composites from filaments, in which the process is free

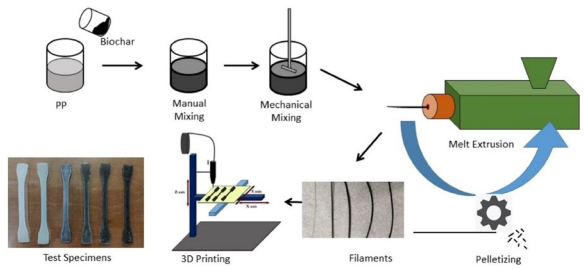


Fig. 1 Common production overview of polymer–biochar filament composite and 3D printing process. From Ref. (Mohammed et al. 2022a). Reprinted from Composites Part C: Open Access, 7, Zaheeruddin Mohammed, Shaik Jeelani, Vijaya Rangari, Effective reinforcement of engineered sustainable biochar carbon for 3D printed polypropylene biocomposites, 4, (2022), with permission from Elsevier

from nozzle clogging, die-swelling, biochar aggregation, etc. The goal of this review is to fill in the gaps of biochar 3D-printed composites, as well as discuss additional biochar processing methods that could pertain to future research as it is currently limited. Biochar processing methods, feedstocks, and post-production biochar treatments are comprehensively reviewed and discussed in terms of their applicability to 3D printing.

2 Biochar-based composites

The role of biochar as a composite filler spans multiple polymer types and applications. Many of the aforementioned applications are significantly influenced by production and processing parameters. In general, the production process involves an initial mixing of the polymer and biochar, as shown in both the mechanical mixing and manual mixing of Fig. 1 (Mohammed et al. 2022a). Afterwards, an extruder is used to produce the filament, with twin screw extruders being popular for mixing composite materials. This particular setup of the twin screws allows for improved mixing due to shear stresses that result from the screws and barrel (Ahmad et al. 2023). This composite filament is then placed into the 3D printer to produce samples (Mohammed et al. 2022a). Shaqour et al. lists four main sections of the 3D printer for operation: the motor to draw the filament through, the barrel to contain the filament, the heating block to melt the polymer, and the nozzle for extrusion (Shaqour et al. 2021).

Furthermore, the goal of biochar fillers is typically focused on either the substitution of harmful environmental materials or the improvement of composite properties. Researchers have used this idea of a biochar filler to study the effects of combining biochar with conventional polymers to form filament composites for applications in 3D printing fields. Diverse polymers, including polyethylene terephthalate (PET), polylactic acid (PLA), high density polyethylene (HDPE), polyurethane (PU), acrylonitrile butadiene styrene (ABS), and polypropylene (PP) have been used with biochar fillers.

2.1 Current trends in biochar/polymer 3D printing: mechanical properties

Though biochar incorporation into 3D-printed composites is relatively new, much of the current research indicates that low concentrations of biochar are the most effective in mechanical reinforcement. For instance, Indrees et al. reported the increase in tensile strength of recycled PET composites with the addition of small concentrations of biochar (0.5 wt.%), from ~ 40 MPa pristine PET to ~ 52 MPa of the composite. The authors attribute this to the interaction of the PET polymer with the porous biochar (0.5 wt.%), whereas higher percentages of biochar decrease tensile strength due to biochar particle aggregation (Idrees et al. 2018). Similarly, Anerao et al. investigated the influence of 3D printing parameters on mechanical properties using PLA/BC composites and determined that small concentrations (1–3 wt.%) of biochar produced a better impact strength than higher percentages (5 wt.%) (Anerao et al. 2023). Vidakis et al. (2024) were able to produce HDPE/biochar filaments for 3D-printed composites, where the biochar was varied between 2 and 10 wt.%. Overall, the authors determined that the two best composites in terms of mechanical properties were the 4 wt.% and 6 wt.% biochar concentrations. For the 6 wt.% composites, the highest tensile strength (25 MPa), toughness (8.5 MJ m^{-3}), and flexural modulus (~ 700 MPa) were found, while 4 wt.% had the highest tensile modulus (95 MPa) and flexural strength (23 MPa) (Vidakis et al. 2024). Further exploration of epoxy resin composites was done by Alhelal et al. by adding biochar (from used coffee grounds) to epoxy resin in a 3D printing process. The biochar produced better mechanical properties in 1% concentrations than those of higher concentrations, in which flexural strength increased by ~ 43% compared to pristine epoxy resin. Additionally, it was noted that 3% biochar concentrations obtained poorer mechanical properties as well as the biochar, in general, having little effect on temperature-induced decomposition (Alhelal et al. 2021). Mayakrishnan et al. developed a 3D-printed BC/PU film (2–10 wt.% of BC) for agricultural practices. The authors collected mechanical properties based on tensile strength, tear strength, penetration resistance, impact resistance, and burst strength and found values of 23–38 MPa, 2.9–4.1 MPa, 22.8–26.7 N, 210–279 g, and 83.6–94.5 kPa, respectively. Those properties were improved with increasing BC content (2–10%). The improvement of mechanical properties was attributed

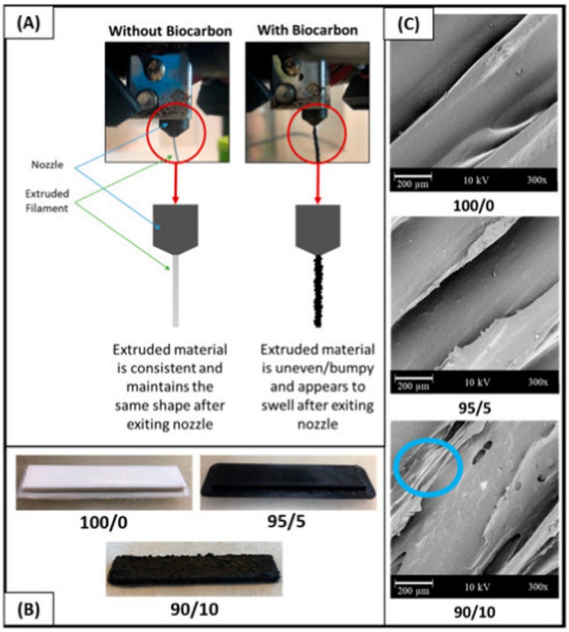


Fig. 2 Inconsistencies in filament caused by biochar aggregation, where **A** shows the filament printing from the nozzle, **B** shows the printed part's surface texture (5 and 10 wt.% biochar concentrations), and **C** shows scanning electron microscopy (SEM) images of the parts. From Ref. Diederichs et al. (2021)

to a range of possible mechanisms including BC promotion of crystallization (affected tensile strength), rigidity (affected tear resistance), and BC dispersion and adhesion to polymer matrix (affected penetration resistance, impact resistance, and burst strength) (Mayakrishnan et al. 2023). In summary, biochar was found to assist in property improvement when typically added in concentrations less than ~ 10 wt.%. However, in order to act as a filler and make a polymer more bio-based, the amount of biochar added to the polymeric matrix must be maximized. On the other hand, at these high concentrations, biochar has been known to aggregate.

Particle aggregation in high concentrations of biochar is a common technical challenge in composite properties and can cause printer nozzle clogging. Diederichs et al. explored the 3D printing of polytrimethylene terephthalate (PTT) and BC composites. Some of their results are shown in Fig. 2, where the textures of three filaments and composites are observed: pristine PTT, 5 wt.% biochar, and 10 wt.% biochar. The authors examined that 10 wt.% of biochar filler caused a rough, inconsistent surface on the printed specimens, as compared to the pristine polymer. They further discussed the impact large BC particle sizes could have on nozzle blockage during extrusion. This was cited as a possible source of decreased mechanical properties, along with poor layer adhesion (Diederichs et al. 2021). Ertane et al. were able to produce PLA/BC (5, 15, and 30 vol.% of biochar) composite filaments and test for tribological properties. According to their study, a 30 vol.% BC composite experienced the highest wear resistance of all tested samples. As BC was added, the wear resistance increased, which was attributed to the improvement of stiffness. However, the nozzle experienced clogging due to the high loading (30 vol.%) of BC in the filament (Ertane et al. 2018). George et al. sought to avoid clogging by sieving the biochar using a 400 mesh size (38 μm), and they experienced no clogging at 220 °C. Additionally, it should be noted that low concentrations of biochar were used (1–10 wt.%) (George et al. 2023).

Rather than only focusing on biochar concentrations, researchers have taken into account the 3D printing processing parameters as well. Though current 3D-printed polymer/biochar composites are limited, research has emerged on potential parameter adjustments in response to quality. Anerao et al. determined biochar had the largest impact of all processing parameter contributions. However, in addition to 3 wt.% of BC, the authors determined that an 80% infill density and 0.3 mm layer thickness were best to maximize tensile properties out of all tested parameters. Similarly, for flexural strength and modulus, the BC content (3 wt.%) and infill density (100%) were found to significantly affect the values. However, a 1 wt.% of BC and an 80% infill density were best for impact strength. They cite the higher density as being important for load transfer, which improves composite mechanical properties. Overall, they determined that infill pattern and raster angle were less important on mechanical property effects. This was done through the Taguchi design that allows for comparisons of which parameter had the most effect on a particular property (Anerao et al. 2023). Khan et al. investigated ABS/BC composites by varying both the biochar content (1–3 wt.%) and infill density (25% and 50%). A higher tensile strength was found for a greater infill density as well as an increase with the addition of small concentrations of BC (Khan et al. 2023). Hassan et al. investigated recycled HDPE and recycled PP composites with biochar and determined the best printing parameters based on mechanical results. The raster angle of 0° aligned each layer in a way that was consistent and improved tensile strength, while a slower printing speed (900 mm min^{-1}) improved layer adhesion (Hassan et al. 2024a). Aside from the biochar itself, it appears that an important factor in 3D printing involved adjusting the infill density, while raster angle adjustments yielded differing conclusions.

Furthermore, a common problem that has emerged across 3D printing research in general is poor layer

adhesion. Because the polymer is deposited layer-by-layer, poor adhesion between each layer can cause poorer mechanical properties, leading many researchers to investigate ways of improving adhesion. Balou et al. attempted to improve layer adhesion by implementing polyethylene terephthalate glycol (PETG) as the 3D printing polymer with hydrochar as a filler. Although hydrochar is slightly different than biochar (with hydrochar requiring a hydrothermal process instead), the material contains a high carbon concentration and is produced from waste biomass. For this polymer, 30% and 50 wt.% of activated carbon derived from hydrochar was added to the PETG matrix to produce higher tensile strength and Young's Modulus compared to neat PETG. The authors cited two reasons for this: first, the interaction of -CH functional groups with the polymer matrix, and second, enhanced crystallinity, which improved filler-matrix adhesion. In terms of the 3D printing aspect, the authors found that the PETG/(30 wt.%) activated carbon composites showed no defects or shrinkage, as occurred in some of the previous 3D printing studies listed (Balou et al. 2023). Mohammed et al. discussed poor adhesion between layers in their study on ultrasonicated biochar filler within a PP matrix. They attributed the poor adhesion to shrinkage upon cooling and attributed the improvement to biochar, which suppressed this by suppressing the volumetric changes (Mohammed et al. 2022a).

While these findings indicate a possible improvement in adhesion using biochar, other authors cite the need for more 3D printing parameterization despite their use of biochar. Vidakis et al. performed scanning electron microscopy in which a more compact and less porous structure was found for the biochar composites compared to pristine HDPE. Therefore, the composites experienced poorer layer adhesion. The authors recommended that different BC concentrations require adjustments to printing parameters (Vidakis et al. 2024). Muhammed et al. investigated PP filaments with plasma-functionalized biochar filler (up to 1 wt.%) for use in 3D-printed filaments and composites. Citing the poor properties of PP that make it inaccessible for printing (e.g., crystallinity), the authors used biochar as a nucleating agent. The authors indicated that the biochar showed improved chemical interaction with the PP matrix due to the plasma treatment, therefore increasing the overall crystallinity. Otherwise, the non-plasma treated biochar only interacted with PP physically. The plasma-treated biochar in a 0.75 wt.% concentration was found to be best in terms of tensile strength (41.5 MPa) compared to pristine PP (21.7 MPa). When printing of the dogbone sample occurred, shrinkage between polymer layers varied the mechanical properties due to inhomogeneous BC dispersion, with the authors acknowledging the need for parameter optimization (Mohammed et al. 2022b). Overall, there is no clear consensus on whether biochar can improve layer adhesion, but rather it depends on the 3D printing parameters and functionalization of the biochar particles.

It appears that throughout much of the work on 3D printed polymer/biochar mechanical properties, indications of poor mechanical properties have been a two-fold problem. First, the printed part exhibits varying adhesion capabilities between each layer. This was due to polymer cooling and shrinkage, resulting in separation between the layers. However, biochar was found to be beneficial in layer adhesion in some cases. Research has suggested an improvement due to biochar incorporation, though additional parameter optimization is still needed. Second, biochar has been found to clog the printer nozzle due to its size, aggregation susceptibility, and inability to melt, especially when added at high concentrations. However, biochar is often investigated as a filler to replace certain expensive polymer matrices and requires a high concentration to make a financial difference. This makes balancing biochar content with printer capabilities an area requiring further investigation.

2.2 Current trends in biochar/polymer 3D printing: thermal properties

In general, because biochar undergoes pyrolysis and contains high concentrations of stable carbon, these composites exhibit great thermal properties. Therefore, it is important to investigate any changes in thermal properties that may occur when added to polymer matrices, including increased melting temperature and degradation effects. Alhelal et al. found that the onset degradation temperatures were similar (325–328 °C) between the neat epoxy, 1 wt.%, and 3 wt.% epoxy/BC composites, though degradation rate changes were less prominent, leading to the conclusion that the changes due to biochar were negligible (Alhelal et al. 2021). However, Umerah et al. found that the biochar/PLA composites were less thermally stable than the pristine polymer and they attribute this phenomenon to the potassium content in the biochar and its degradation of the PLA itself. Their TGA results can be seen in Fig. 3, where an earlier degradation temperature appears as biochar content is added to the polymer matrix (Arrigo et al. 2020; Umerah et al. 2020). Nevertheless, the authors stated that a maximum processing temperature of 250 °C can be used without polymer degradation occurring (Umerah et al. 2020). Vidakis et al. investigated HDPE/biochar composites for degradation using TGA and determined similar degradation temperatures during the stage of significant polymer loss (Fig. 4) (Vidakis et al. 2024). Overall, despite biochar

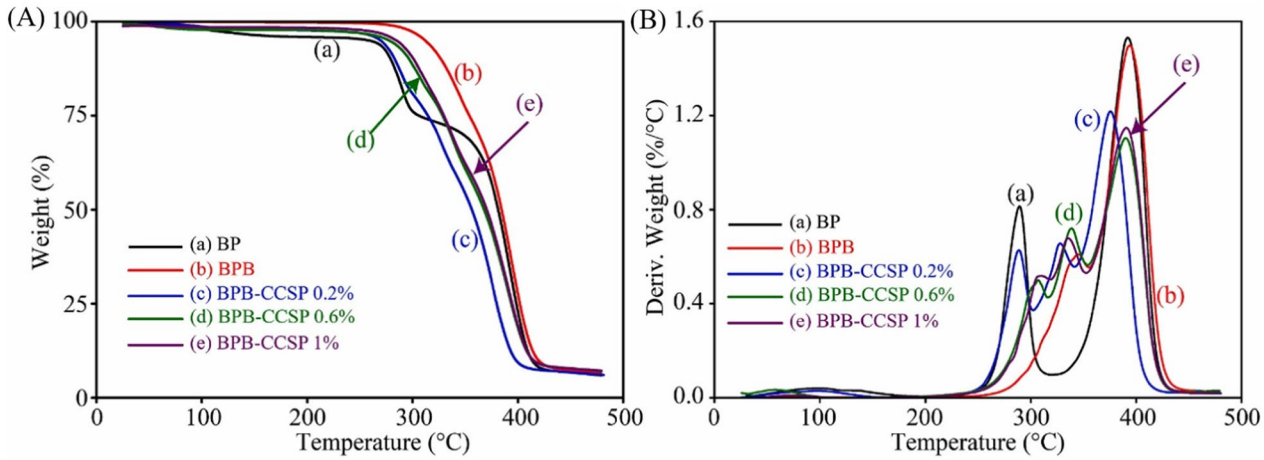


Fig. 3 Thermogravimetric analysis (TGA) (A) and derivative weights (B) of Bioplast polymer (BP), Bioplast-PLA blend (BPB), and BPB with coconut shell powder biochar (CCSP) in varying concentrations (0.2, 0.6, 1%). From Ref. (Umerah et al. 2020). Reprinted from Composites Part B: Engineering, 202, Chibu O. Umerah, Deepa Kodali, Sydnei Head, Shaik Jeelani, Vijaya K. Rangari, Synthesis of carbon from waste coconutshell and their application as filler in bioplast polymer filaments for 3D printing, 4, (2020), with permission from Elsevier

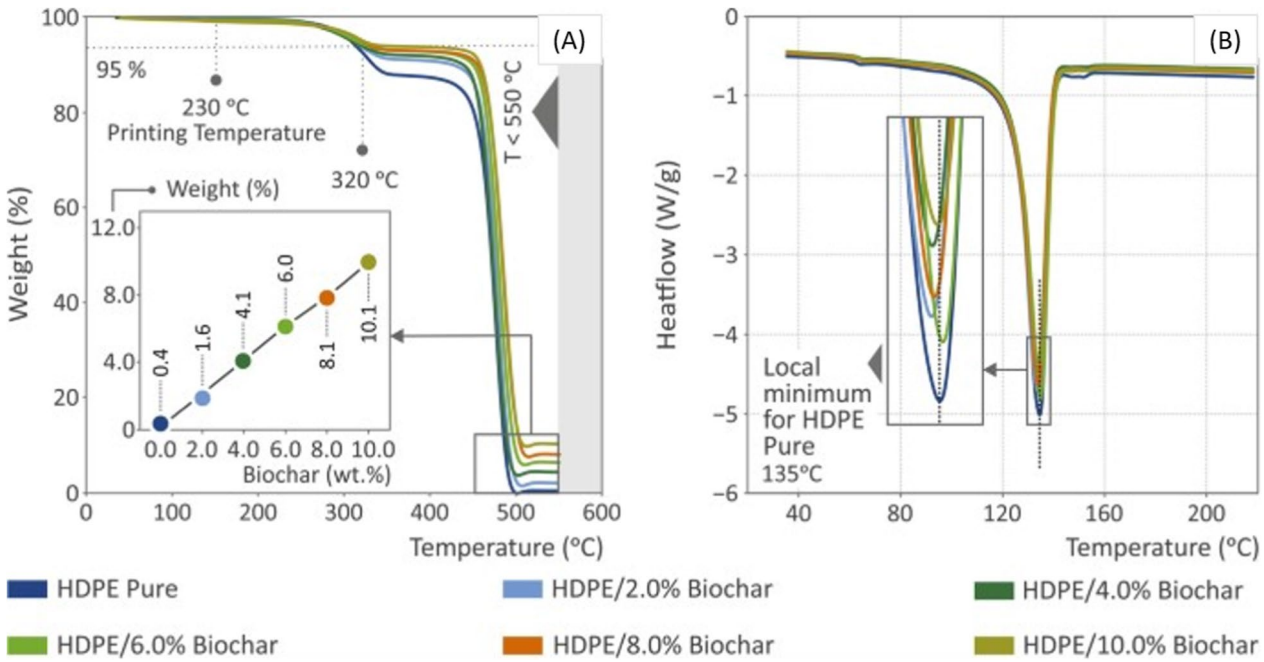


Fig. 4 TGA (**A**) and DSC (**B**) data for HDPE composites with varying amounts of biochar. From Ref. Vidakis et al. (2024)

content possibly influencing polymer degradation as in the case of the potassium content, the composites are able to maintain consistent onset degradation temperatures at low concentrations after replacing portions of the polymer matrix with the biochar filler.

In addition to the degradation temperature above, the melting temperature of the composites is commonly examined as a thermal property. Mohammed et al. determined that plasma-treated biochar within a PP matrix increased the melting temperature and attributed it to the chemical bonding that occurred between the biochar and polymer as a result of plasma functionalization. In terms of crystallization, the chemical bonding occurred as covalent bonds and increased the energy needed for crystallization (Mohammed et al. 2022b). In another study, Mohammed et al. reported that 1–10

wt.% addition of biochar could slightly increase the melting temperature of PP composites from 159.72°C (pristine) to ~ 163°C (composite) (Mohammed et al. 2022a). It appears that biochar could increase the melting temperature; therefore, printing parameters must be adjusted accordingly. Temperatures can either be adjusted across varying composite trials to match biochar concentration or be held constant across trials according to the control polymer. To further investigate optimum printing parameters, George et al. investigated not only the melting temperature of the composites but their onset melting temperature as well. They determined that this temperature was an indication of the nozzle temperature, in which 220 °C was chosen for PLA/PBAT/biochar composites. This temperature was chosen to avoid clogging and degradation (George et al. 2023). Notably, melting only occurs within the polymer rather than the biochar filler (Ertane et al. 2018). This is a significant part of nozzle clogging. Diederichs et al. described swelling of the filament during extrusion from the printer nozzle as well as the inconsistent rough surface of the printed part. They determined that 5 wt.% BC was better for printability than the addition of 10 wt.% biochar (Diederichs et al. 2021).

As previously discussed, layer adhesion can lead to mechanical failure but is influenced by temperature, making it a thermal property. Mohammed et al. cite that in order to improve layer adhesion, the previous layer temperature must remain above crystallization but below melting temperature (Hertle et al. 2016; Mohammed et al. 2022a). The authors further attribute their addition of biochar to the polymer matrix as disrupting the shrinkage that normally occurs without the filler, though this is for the filament rather than the printed sample. This is due to the chain movement restriction caused by the biochar addition (Mohammed et al. 2022a). Similarly, Bute et al. reported that in common FDM polymers (PLA, ABS, PP, etc., without biochar), thermal shrinkage occurred after deposition in the printing plane whereas expansion occurred within the extrusion plane (Bute et al. 2023). Balou et al. further explored the use of biochar composites and determined improved stability and restrained polymer chain movement owing to both fillers. They determined that no shrinkage or defects occurred using the activated carbon (biochar) at 30 wt.% within a polyethylene terephthalate glycol matrix (Balou et al. 2023). However, this is considered a high biochar concentration and previously discussed research has shown difficulties with nozzle clogging and mechanical failures caused by aggregation.

Overall, thermal properties play a significant role in influencing the two most common 3D printing difficulties: biochar nozzle clogging and polymer layer adhesion. Biochar appeared to influence the melting temperature of the composites (Hassan et al. 2024b). This should be considered when choosing the nozzle temperature, as it is influenced by the onset melting temperature (George et al. 2023). Therefore, it is also important to carefully choose the biochar concentration according to the intended application to prevent a blockage in the nozzle because of its inability to melt (Ertane et al. 2018). This causes difficulties in the processability of the polymer. However, conflicting reports remain regarding the effect of biochar on thermal shrinkage, requiring additional investigations. Therefore, it is important to carefully select the biochar concentrations and parameters for a particular application.

2.3 Current trends in biochar/polymer 3D printing: additional properties

Although many works have focused on mechanical and thermal characteristics, other studies have explored additional properties in which the biochar is tailored for a specific application. These properties are valuable in biochar and, as such, tested according to a specialized application. For example, Silva et al. produced 3D-printed (though not FDM) alginate-based composite hydrogels with varying concentrations of biochar. Although mechanical characterization was not done, contaminant absorbency tests were performed. The 10 wt.% biochar samples were found to have the highest absorbency due to porosity induced by the biochar addition. An improvement of 48–58% in absorbency for tested water contaminants was found compared to the pristine alginate hydrogel (Silva et al. 2023).

Instead of contaminants, Mayakrishnan et al. expanded on composite characterization by testing water absorbance for biochar-based mulching films. Overall, the authors found no significant changes in water absorbance between the pristine thermoplastic polyurethane films and those with biochar (2–10 wt.%). No significant defects were observed as a result of the 3D printing process, supporting the polymer composite stability (Mayakrishnan et al. 2023). Mayakrishnan et al. also investigated oxygen transmission through composite films. With the addition of biochar, the oxygen transmission rate was decreased, with the pristine polymer being 1765.45 cc m^{-2} day^{-1} atm^{-1} and the 10 wt.% biochar composite being 1011.34 cc m^{-2} day^{-1} atm^{-1} (Mayakrishnan et al. 2023).

Diederichs et al. investigated the rheology of molten composites in order to determine their effects on 3D printing. They determined that the polymer composite experienced shear thinning, which changed with biochar content, thereby influencing the nozzle pressure during printing (Diederichs et al. 2021). Baniasadi et al. explored

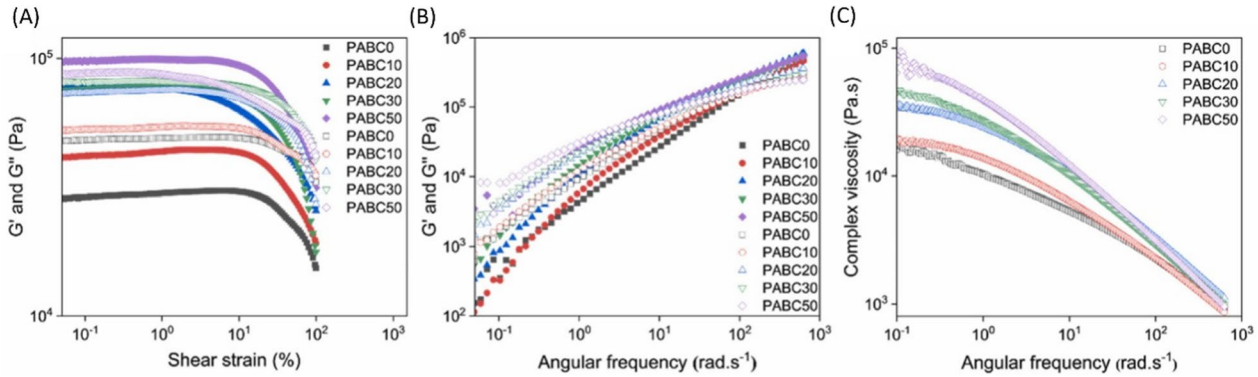


Fig. 5 Rheological data collected for polyamide 11 (PABC0) and its biochar-based composites (PABC10, PABC20, PABC30, PABC50). The authors define shear storage modulus (*G'*) by the solid symbols, while the shear loss modulus (*G"*) is represented by the unfilled symbols. From Ref. Baniasadi et al. (2023)

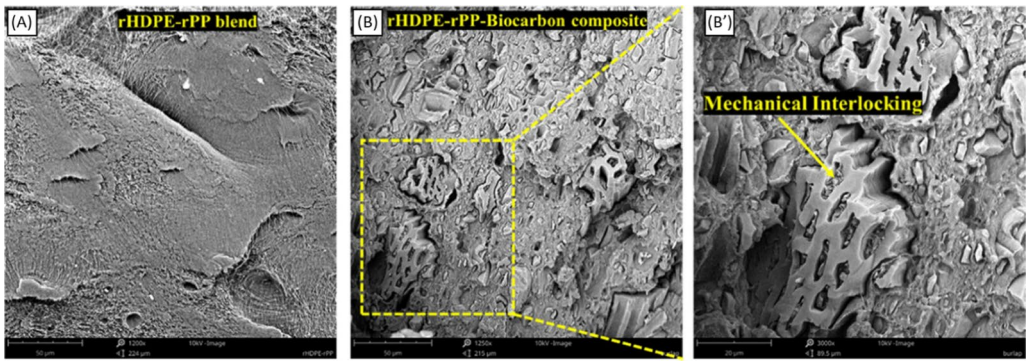


Fig. 6 SEM images (**A**, **B**, and **B'**) demonstrate the interlocking effect that occurs between porous biochar and polymer. Adapted with permission from (Hassan et al. 2024a). Copyright 2024 American Chemical Society

rheology and discussed the importance of biochar dispersion in filaments. Their results (Fig. 5) showed that higher frequencies caused a decrease in the viscosity of the composites, known as shear-thinning. They explained that shear-thinning is needed for the filament to easily flow out of the nozzle to limit defects. The molecules are able to more easily move past each other, leading to improved movement. Their success in producing such a filament was attributed to the in situ polymerization of polyamide 11 and biochar into a composite rather than extrusion techniques to combine the two (Baniasadi et al. 2023).

In a work by George et al., biochar 3D-printed composites were produced to study their antistatic properties. The authors were able to improve the polymer blend adhesion (PLA and polybutylene adipate-co-terephthalate) and decrease the surface resistivity. They cite the change in resistivity as occurring due to the sp^2 hybridized carbon content, where the coconut shell-derived biochar has few impurities and high carbon content. Additionally, adhesion was improved by biochar nucleation when added to the polymer blend (George et al. 2023).

3 Biochar production and properties

The surface characteristics, porosity and composition of biochar can potentially influence composite properties, as well as its printability. Although polymer/BC composites typically do not react chemically (unless modified), physical interactions can occur. The molten polymer can be deposited within the porous surface of the biochar, solidifying as it cools and interlocking the two materials, as shown in the SEM images from Hassan et al. (Fig. 6). The authors attribute the decrease in tensile properties to weakened polymer adhesion and variations in particle size. However, they attribute increases in flexural properties to the deposition of the polymer into biochar pores. For example, the authors indicate that during flexural testing, the biochar provides structural support for the composite when undergoing compression (Hassan et al. 2024a). In particular, the study by Hassan et al. highlights that it is important to consider surface morphology and

biochar interactions with the molten polymer, thereby placing emphasis on biochar surface formation during production. Biochar properties can be tuned using two main factors resulting from the production process: the pyrolysis conditions and feedstock content. Pyrolysis was selected as the main focus of the production process due to its high biochar yield. Adjusting these parameters can significantly tune biochar, leading to influences on printing properties. This section begins with a discussion of previous studies on the pyrolysis process and continues with feedstock options for obtaining varying biochar compositions and properties. Significant attention is given to waste materials as potential feedstocks. A summary of feedstocks and temperatures is provided in Table 2. This table provides an overview of what properties can be estimated for a particular feedstock.

3.1 Pyrolysis conditions

Pyrolysis is a widely used method of heating organic materials at temperatures between 300 and 900 ℃ within a limited oxygen environment, thereby breaking down biomass into gas, liquid, and solid products like biochar. This method can be categorized as either slow or fast pyrolysis, with yields depending on the applied heating rates, processing temperatures, and residence times. These parameters result in different product distributions (syngas, liquid bio-oil, and solid biochar). Slow pyrolysis is carried out at the temperature range of 300–700 °C with a heating rate of 0.1–1 ℃ s^{-1} and is typically used for biochar production because of its relatively high char yield compared to other methods. Its counterpart, fast pyrolysis (400–800 °C), is an exceptionally fast process with a short residence period of < 2 s (Pahnila et al. 2023). In addition, microwave pyrolysis (heating biomass using microwave radiation) has several advantages, including a short processing time, increased efficiency, and a non-contact manner (Wang et al. 2018). With this method, biochar with higher carbon content and higher calorific value has been obtained, compared to the one via conventional pyrolysis (Said et al. 2022). Co-pyrolysis can be applied to the mixture of two or more materials or chemicals as catalysts, improving the physical and chemical properties of the produced biochar (Chen et al. 2022; Liu et al. 2021; Peng et al. 2022). For instance, Peng et al. reported that co-pyrolysis of industrial sludge and rice straw produced biochar with increased specific surface area and a more developed pore structure than pyrolysis using industrial sludge alone (Peng et al. 2022).

When adjusting pyrolysis parameters, temperature is often the focus of the overall biochar outcome. In terms of yield, the highest percentage of biochar is produced at lower temperatures (Dhar et al. 2022; Vieira et al. 2020; Wang et al. 2019). One significant example is found in a study by Wang et al., in which ~ 80% yield was generated at 300 °C. At higher temperatures (400–700 °C), the yield ranges from 60% to 73% for textile dyeing sludge, with percentages decreasing with increasing temperatures (Wang et al. 2019). A high biochar yield can be beneficial

Table 2 Feedstocks and their resulting biochar properties

Feedstock	Temperature (°C)	Yield (wt.%)	Surface area ($m^2\ g^{-1}$)	Volatile matter (wt.%)	Fixed carbon (wt.%)	Ash content (wt.%)	Reference
Textile dying sludge	300–700	60–81	21–66	–	–	63–78	Wang et al. (2019)
Rice husk	300–500	33–38	–	7–47	30.5–60	17–32	Vieira et al. (2020)
	300–700	35–55	–	–	–	21–35	Nwajiaku et al. (2018)
	450	~ 31	–	~ 17	~ 55	~ 23	Chaturvedi et al. (2021)
Coconut fiber	350–600	29–48	8–296	16.5–58.5	35–71	6–12	Dhar et al. (2022)
Woody yard waste	450–950	23–33	1–310	–	–	1–9	He et al. (2021)
Pine wood	300–700	15–37	4–380	–	–	0.5–4	Usevičiūtė and Baltrėnaitė-Gedienė (2021)
Pine bark		14–55	1–420			3–5	
Birch wood		11–31	2–345			0.5–3	
Birch bark		16–49	0.5–380			2–7	
Hemp		11–40	2–392			3–32	
Grapevine cane	400–700	–	183–516	–	–	–	Marshall et al. (2019)
Grapevine stalks	400–700	–	200–560	–	–	–	
Sugarcane bagasse	350–700	22–27	–	–	–	11–12	Nwajiaku et al. (2018)
Sugarcane trash	450	~ 31	–	~ 15	~ 61	~ 20	Chaturvedi et al. (2021)
Forestry waste (pine, lantana, eucalyptus)	450	29–36	–	17–37	50–70	7–9	Chaturvedi et al. (2021)
Vegetable	300–600	16–45	1–10	16–73	19–70	–	Pradhan et al. (2020)
Animal-based	200–500	–	0.01–2	45–82	0–35	2–55	Fu et al. (2019)
Grain-based	200–500	–	0.04–1.2	40–90	6–50	0.5–7	

to the composite-making process, as an in-demand application would need a sustainable resource to draw from for production. Moreover, the demand for biochar would produce bio-oil as a result of pyrolysis, further promoting the utilization of these fuels for a more renewable energy source. Additional tests on the effect of pyrolysis temperature were done to examine the specific surface area of biochar. Typically, at higher pyrolysis temperatures, the surface area of the biochar increases owing to volatile matter release (Dhar et al. 2022; Tomczyk et al. 2020; Wang et al. 2019). This additional surface area often results in increased porosity. Surface area and porosity are significant in their interactions with polymer matrices as the polymer can deposit itself within the pore, interlocking the two matrices with the potential for improved mechanical properties (Aup-Ngoen and Noipitak 2020). For example, it was found that biomass utilizing coconut fibers at 350 °C produced a specific surface area of ~ 8 $m^2 g^{-1}$ compared with ~ 296 $m^2 g^{-1}$ at 550 °C (Dhar et al. 2022). Further results showed a ~ 24 $m^2 g^{-1}$ surface area at 300 °C compared to the higher temperature of 700 °C at ~ 65 $m^2 g^{-1}$ (Wang et al. 2019). Umerah et al. utilized coconut shell waste to fabricate 3D printing filaments using biochar pyrolyzed at 800 °C, resulting in an increase of tensile strength, though Brunauer–Emmett–Teller (BET) data for the surface area were not reported (Umerah et al. 2020).

3.2 Feedstock choice

While pyrolysis itself can affect biochar properties, feedstock selection has become just as important. However, controversy has ensued over the years regarding whether agricultural crops should be used in pyrolysis or to feed communities. Furthermore, much of the focus on feedstocks has shifted toward waste, including agricultural, food, and industrial wastes. Many studies have examined the potential properties of this biochar feedstock. Such feedstocks include rice husk (Chaturvedi et al. 2021; Nwajiaku et al. 2018; Vieira et al. 2020), sugarcane bagasse (Chaturvedi et al. 2021; Nwajiaku et al. 2018), and straw (rice and maize) (Chaturvedi et al. 2021). Both Nwajiaku et al. and Chaturvedi et al. determined that sugarcane bagasse and rice husk waste-derived biochar could enhance soil properties due to available nutrients and pH control (Chaturvedi et al. 2021; Nwajiaku et al. 2018). Chaturvedi et al. pyrolyzed forestry waste (including eucalyptus, lantana, and pine) and determined the resulting biochar to have a low ash content coupled with an elevated fixed carbon content from a wood-based feedstock (Chaturvedi et al. 2021). Ash content is significant for polymer composites as too high a content could cause aggregation of the biochar particles. It was determined that lignin-rich biomass could limit ash content (Li et al. 2023a). Furthermore, aggregation in composites not only holds the risk for poor mechanical properties, but also nozzle clogging. Lignin-based biomass may assist in preventing nozzle clogging as well as improved composites. Table 2 lists the various biomass types and associated properties to assist in feedstock choice.

Food waste is another popular option for research on pyrolysis feedstocks as it takes unwanted, leftover portions of food and provides new uses. Such studies have utilized vegetable-based (Pradhan et al. 2020), animal-based (Fu et al. 2019), and grain-based (Fu et al. 2019) foods. The authors concluded that biochar from a mixture of various vegetable wastes was comparable to vegetable waste of a singular vegetable type. Furthermore, this is best for realistic household and industry disposal methods (Pradhan et al. 2020). A comparison of the animal products (eggshells, fish) and the grain-based foods (breadcrumbs, rice) revealed that fixed carbon, an indicator of higher biochar yield, was highest in the breadcrumbs and rice waste, (10–50% and 7–48%, respectively) but lowest in eggshells in which a percentage of zero fixed carbon was reported. The authors suggested food waste with a high carbohydrate content for better biochar production yield. One notable drawback was the low biochar surface area due to the lack of lignin and cellulose contained in the waste (Fu et al. 2019). This could have a significant impact on composite properties as surface area can significantly influence polymer–biochar interactions. Biochar has been cited for improved flexural strength, attributed to its high surface area facilitating polymer interactions (Das et al. 2016; Hassan et al. 2024a).

Although more plant-based approaches exist, industry waste can be seen as a possible source of biochar in the pyrolysis process. Ghodke et al. studied sewage sludge from wastewater and identified both bio-oil and biochar properties in comparison with those of the pristine feedstock. The biochar was found to have a low moisture content (~ 2 wt.%) as well as a significant ash content of ~ 60 wt.% (Ghodke et al. 2021). While ash content was previously discussed in the context of aggregation, its removal has been associated with improved flexural modulus as well (Zhang et al. 2020).

The descriptions above provide insight into the current studies based on biochar production parameters with a focus on potential FDM processing. Based on these, it appears that biochar is highly tailorable to a specific application, and its resulting properties have the potential for influencing 3D printing. It is best for researchers to appropriately choose the correct feedstock before pyrolysis, as the target application will require different demands of the biochar. It may be best to choose a biochar feedstock (for FDM processing) containing high amounts of lignin, such as woody biomass waste, to

reduce ash content and provide more surface area for polymer interactions. This could improve mechanical properties as well as assist in polymer flow through the printer. Additionally, the pyrolysis temperature should be adjusted according to the surface area and porosity results.

4 Biochar treatment and modification

4.1 Biochar physical modification

The physical treatment of biochar plays a critical role in the mechanical performance of 3D printing as many applications have high mechanical property requirements (e.g., 3D-printed car, 3D-printed house). Appropriate physical treatment of biochar can optimize the particle size, surface area, and generation of functional groups. For example, Mozrall et al. explored the effects of different processing methods on biochar particle size, and determined that particle sizes ~ 150 µm or less showed improved tensile properties compared to those at 250 µm. The authors attribute the improvement in mechanical properties to an increase in the biochar surface area (Mozrall et al. 2023). Although not biochar, poplar fibers have been used in composites for large-scale 3D printing to prepare poplar-PLA composites. It was found that as poplar particle size (< 180, 180–425, 425–850, and 850–2360 µm) decreased, tensile strength increased, which was attributed to improved particle dispersion and access to particle surface porosity. This result may indicate that if the biochar particle size is small (e.g., < 180 µm), the biochar-polymer composite may also have a high tensile strength (Zhao et al. 2019). In addition, another review mentioned that the acceptable particle size statistic depends on the nozzle diameter of the 3D printer. Benchtop 3D printers recommend the additive be milled to a micrometer scale to avoid nozzle clogging (Bhagia et al. 2021). Additionally, George et al. noted that a 400 (38 µm particle size and smaller) mesh size could help prevent the clogging of the printer during the biochar composite extrusion (George et al. 2023). Thus, particle size and surface area of biochar are crucial for composite implementation, and it has been determined that physical treatments can modify the biochar internally and morphologically (Chatterjee et al. 2018).

To achieve the desired particle size, surface area, and functionalization of biochar, some representative methods can include ultrasound irradiation, grinding, and ball milling. For example, biochar can be treated under different ultrasound irradiation durations such as 0.5, 1, and 3 min (Chatterjee et al. 2018). Ultrasound can improve the microporous surface area of biochar from ~ 312 to ~ 354 $m^2 g^{-1}$. One possible reason is that the micro-jets created during sonication affect the biochar surface and develop a more porous structure (Chatterjee et al. 2018). Mohammed et al. used ultrasonication as a method of biochar processing to improve biochar incorporation into 3D-printed composites. They found an increase in surface area of 185.08 $m^2 g^{-1}$ for the ultrasonicated biochar from the unprocessed biochar at 2.99 $m^2 g^{-1}$, in which the authors attributed it to both decreased particle size and surface nanofeature developments as a result of ball milling. The authors noted changes in crystallization that improved thermal stability, as well as improved printing ability by decreasing printer die blockage (Mohammed et al. 2022a). Gupta and Kua ground biochar using a manual process, which was conducted with a mortar and pestle to break down the large chunks into powder form. The overall size of the ground biochar ranged from 2 to 100 µm, whereas the macropores on the surface of the ground biochar were 10–20 µm in size. The external surface area was ~ 56 $m^2 g^{-1}$. In contrast, the authors explored ball milling as well and determined that the biochar had smaller particle sizes (100 nm–2 µm) and a higher external surface area (~ 76 $m^2 g^{-1}$). The authors explain that the macroporous structure was destroyed during ball milling, with few changes in microporous structure occurring (Gupta and Kua 2019).

Ball milling is a physical treatment method for biochar modification. Ball milling is a top-down method for the production of nanoparticles in which mechanical force is used to reduce particle size (Naghdi et al. 2017). It has been determined that the precision of 3D printing can be influenced by the size of the particles (Diederichs et al. 2021). In addition, ball milling has the potential for green, reproducible, and large-scale production of different classes of nanoparticles. Naghdi et al. used a ball milling method to process biochar in a planetary ball mill. During the ball milling process, several factors were studied, including the time (1.6–8.4 h), biochar mass (5–15 g), and milling speed (516–634 rpm). Results showed that varying biochar weight had a minor effect on the particle size at a shorter milling time. The milling time and speed played a crucial role. After ball milling, the biochar exhibited a higher surface area and cumulative pore volume than raw biochar due to the development of its microporous features. In addition, Naghdi et al. utilized cryogenic conditioning of biochar samples before ball milling to produce nano-sized biochar. Furthermore, cryogenic conditioning is helpful in preventing nanoparticle agglomeration (Naghdi et al. 2017). Combined, these factors may assist in nozzle clogging for 3D printing by reducing aggregation and particle size. Aggregation becomes a main concern when it is implemented into the polymer material, as material variations occur based on the degree of dispersion (Yasim-Anuar et al. 2022). In cement composites, biochar has been found to aggregate according to van der Waals forces (Zhao et al. 2024).

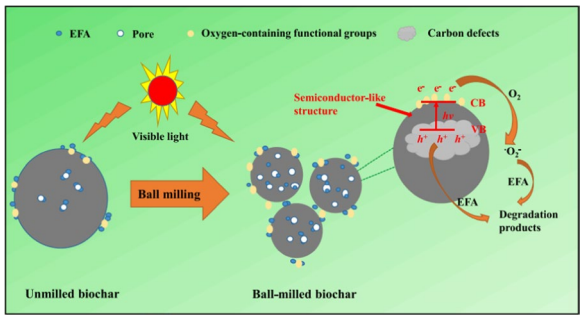


Fig. 7 The proposed ball milling mechanism for obtaining smaller biochar particles, from Ref. (Xiao et al. 2020). Reprinted from Chemical Engineering Journal, 384, Yao Xiao, Honghong Lyu, Jingchun Tang, Kun Wang, Hongwen Sun, Effects of ball milling on the photochemistry of biochar: Enrofloxacin degradation and possible mechanisms, 10, Copyright (2020), with permission from Elsevier. The legend corresponds to enrofloxacin (EFA), pores, functional groups, and defects on particle surface

Therefore, it is important to emphasize production and post-processing characteristics of biochar to maximize dispersion when implemented into polymer matrices.

Although research is limited, it should be noted that ball material itself can influence milling outcomes. A review by Stolle et al. suggests the usefulness of both higher ball material density and an emphasis on reaction classifications that occur as a result of the milling. It is hoped that these two parameters will produce greater yields in terms of samples requiring a chemical reaction in the process of ball milling, although biochar was not mentioned as an example within the article. This may be due to the increased energy required for these two parameters (Stolle et al. 2011).

During physical processing of the biochar, chemical changes that can affect the material could occur. Ball milling can improve the open tips and surface area of carbon-based materials, thereby enabling functional group formation at the open ends. Xiao et al. investigated the effect of ball milling on the physicochemical properties of biochar. After ball milling, the concentrations of lactonic, carboxyl, and phenolic hydroxyl groups increased. The ball-milled biochar was found to have more oxygen-containing functional groups (e.g., O–C=O) as well as carbon defects. This caused a photocatalytic effect in which electrons were moved from carbon to oxygen-based functional groups, promoting EFA degradation. The proposed ball milling mechanism is illustrated in Fig. 7 (Xiao et al. 2020). Vidakis et al. found that their biochar (later used for 3D printing composites) contained a high amount of oxygen-containing functional groups. This is significant as it could mean a hydrophilic material with high degradability due to its reactivity (Vidakis et al. 2023). Furthermore, ball milling increased the external and internal surface areas of biochar by reducing particle size and opening the internal pore network, respectively. Meanwhile, oxygen-containing functional groups were also introduced to the surface of biochar, which can improve the sorption capacity (Lyu et al. 2018). Fourier Transform Infrared Spectroscopy (FTIR) in a study by Balou et al. revealed that the functional groups of activated carbon and PETG bonded and reacted chemically, as indicated by the ~ 1404 cm^{-1} band, where the methyl groups showed changes from the pristine polymer (Balou et al. 2023). This could enhance the FDM process by not only improving the biochar-polymer interlocking mechanism due to compatibility but also by factoring in a chemical mechanism to improve the composite as well.

Similarly, Lopez-Tenllado et al. modified biochar using a high-energy planetary ball mill with ZrO_2 balls and ZrO_2 grinding bowls. The surface area and pore volume measured by N_2 adsorption increased after ball milling, which was attributed to the development of the microporous structure through the opening of obstructed pores. In addition, the total number of accessible phenolic, carboxylic, and lactonic groups on the biochar surface was found to increase. Furthermore, the milling time (0.05 to 12 h) was varied, indicating that ball milling the biochar sample for just 0.05 h could cause a significant increase in porosity. When the biochar was ball milled with the addition of heptane, the agglomeration of biochar was reduced (Lopez-Tenllado et al. 2021). Aggregation is a significant factor in the failure of mechanical testing of BC/polymer composites. This is often cited because it disrupts matrix continuity and, as previously discussed, can affect printability.

In addition, Xu et al. investigated biochar ball milling under different atmospheres (vacuum, N_2, and air) (Xu et al. 2021). An oxygen-limited atmosphere (vacuum and N_2) was more favorable for reducing biochar size than an air atmosphere. The formation of O moieties on the biochar was mainly due to the oxidation process during ball milling and the binding of heteroatoms to the carbon structure. This suggests that an oxygen-limited atmosphere (vacuum or N_2) during ball milling inhibits the formation of O moieties on biochar (Xu et al. 2021). As previously mentioned, oxygen can affect the reactivity of the biochar, and ultimately its wettability and stability. Figure 8 shows the SEM images of pristine and ball-milled biochar. The pristine biochar exhibited an irregularly shaped bulk with a particle size > 100 μm, which was ball-milled to small-sized particles (Xu et al. 2021).

Wet ball milling is a nonequilibrium processing method that can be used to tailor the nitrogen doping level using $NH_3{\cdot}H_2O$. Wan et al. utilized a wet ball milling method to modify biochar, and the resulting material exhibited

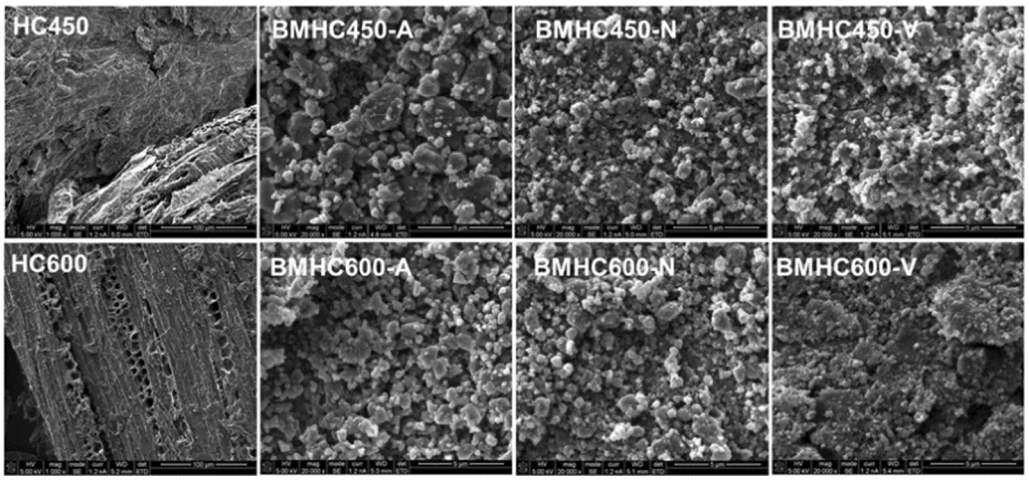


Fig. 8 SEM images of pristine biochar and ball-milled biochar under different atmospheres (HC450: biochar obtained from heating at 450 °C, HC600: biochar obtained from heating at 600 °C, BM: ball milled, A: air atmosphere, N: N_2 atmosphere, V: vacuum atmosphere) from Ref. Xu et al. (2021). Reprinted from Chemical Engineering Journal, 413, Xiaoyun Xu, Zibo Xu, Jinsheng Huang, Bin Gao, Ling Zhao, Hao Qiu, Xinde Cao, Sorption of reactive red by biochars ball milled in different atmospheres: Co-effect of surface morphology and functional groups, 3, Copyright (2021), with permission from Elsevier

a smaller average particle size, smoother surface, as well as decreases in both macroporosity and microporosity compared to pristine biochar. The decrease in porosity is cited as being due to a combination of structure collapse and excessive nitrogen. After ball milling, samples were annealed at different temperatures (500–800 °C), where temperature influences increased surface area compared to the ball-milled biochar that was not annealed. In addition, wet ball milling introduced amino groups on the biochar surface through the use of $NH_3{\cdot}H_2O$ during milling (Wan et al. 2021). Goh et al. investigated the wet ball milling of pine-based biochar. The process consisted of ball milling with a 1:1:40 (biochar: ethanol: stainless steel ball) weight ratio at 10 min intervals. The wet ball milling for 50 min yielded an average particle size of ~ 0.24 µm for the resulting biochar (produced at 550 °C) (Goh et al. 2021).

In summary, ultrasound irradiation, grinding, and ball milling have been used for the physical treatment of biochar. Among these treatments, ball milling has emerged as a prevalent approach for generating smaller biochar particles with a higher surface area due to micropore development. There are various types of ball milling, such as vibration, attrition, tumbler, and planetary ball milling, though Kumar et al. suggested that planetary ball mill is well-suited for lab experiments as it is compact and easy to use (Kumar et al. 2020). To produce biochar well-suited for 3D printing, it may be most practical to utilize the planetary ball mill and carefully consider the resulting particle size and functional groups on the biochar surface. It may be best to run the ball mill in a nitrogen atmosphere to limit the oxygen moieties, as suggested by Xu et al. (2021). However, the addition of oxygen to biochar may be more desired depending on polymer compatibility. Without these considerations, researchers may run the risk of printer clogging and poor polymer–biochar compatibility.

4.2 Biochar chemical modification

Chemical modifications of biochar offer many opportunities for tailoring the biochar to a particular usage. Various chemical treatment methods have been studied to modify or improve biochar properties for its intended applications, including biochar composites for 3D printing. Acidic and alkaline treatments are commonly used to improve textural parameters (e.g. pore volume, pore size, surface area, etc.), surface chemistry, and adsorption capacity of biochar. Acid treatment can remove impurities from biochar, modify its surface functional groups, and develop micropores (Tomczyk et al. 2022). Industrial-grade inorganic acids, including hydrochloric (HCl), sulfuric (H_2SO_4), phosphoric (H_3PO_4), and nitric (HNO_3) acids are usually applied in biochar modification, which reduces cost. For example, Liu et al. treated biochar produced from walnut shells using H_2SO_4 and H_3PO_4 separately for 8 h and compared the effects of the acid treatments (Liu et al. 2020). The acid treatments increased the internal surface area and total pore volume (TPV) of walnut shell biochar. The treatments resulted in opposite effects on the average pore size (APS) of biochar, with an increased APS (from ~ 2.67 to ~ 3.26 nm) observed after the H_2SO_4 treatment and a decreased

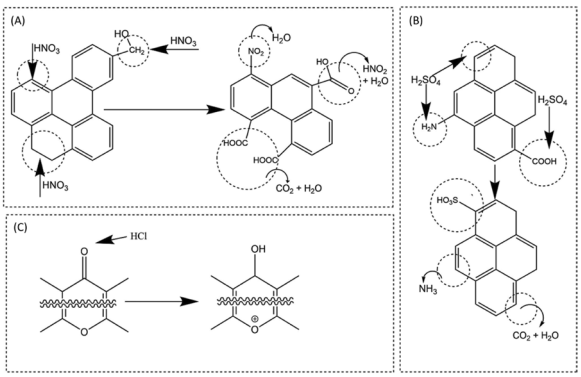


Fig. 9 Chemical reaction mechanism for nitric acid (**A**), sulfuric acid (**B**), and hydrochloric acid (**C**) treatments. From Ref. Peiris et al. (2019)

APS (from ~ 2.67 to ~ 2.43 nm) due to the modification with H_3PO_4. However, inorganic acid treatments can also deteriorate the textural parameters of biochar, decreasing the likelihood of polymer interactions with biochar pores and surface area. It has been reported that pine wood biochar had a decreased surface area and total pore volume after treatment with H_2SO_4 or H_3PO_4. H_2SO_4 treatment increased the value of APS from ~ 3.45 to ~ 4.34 nm, and the treatment with H_3PO_4 reduced this parameter to ~ 3.19 nm for the pine wood biochar (Liu et al. 2020).

In another study, Peiris et al. demonstrated that the acid modification of biochar produced from tea waste with H_2SO_4, HNO_3, and HCl varied BET surface area, TPV and APS, depending on pyrolysis temperature and treatment type. Figure 9 shows the proposed chemical mechanism for the acidic treatments, with the authors stating the addition of oxygenated surface functional groups through the ring opening of nitric acid (Fig. 9A). Furthermore, sulfuric acid treatment (Fig. 9B) occurred due to decarboxylation during the hydrochloric acid treatment (Fig. 9C) functions according to the oxygenated functional groups (Peiris et al. 2019). In addition, inorganic acids (e.g., HCl, HNO_3, H_2SO_4) can act as oxidizing agents, leading to surface oxidation with an increased content of oxygen-containing groups on the biochar surface and a higher molar O/C ratio of biochar. As previously discussed, oxygen functional groups were found to impact polymer compatibility due to their hydrophilic properties, but this could also indicate susceptibility to degradation through reactivity (Vidakis et al. 2023). The effects of acid treatment on biochar properties vary depending on the biochar feedstock, treatment conditions, as well as acid type employed.

Alkali treatment has also been explored to modify biochar properties (e.g., KOH, NaOH, ammonium hydroxide-NH_4OH). The metal hydroxide treatment can increase BET surface area, porosity, and oxygenated functional group content. Huang et al. used KOH to treat poplar sawdust biochar and reported improved textural parameters and increased oxygenation of its surface groups. The treated biochar had increased BET (from ~ 12 to ~ 107 m^2 g^{-1}), TPV (from ~ 0.004 to ~ 0.006 cm^3 g^{-1}) and atomic O/C ratio (from ~ 0.10 to ~ 0.12) (Huang et al. 2017). El-Nemr et al. reported that treatment of biochar produced from *Pisum sativum* peel with NH_4OH improved the biochar's textural parameters and increased the number of functional groups, especially hydroxyl groups, on its surface (El-Nemr et al. 2020). The ammonium hydroxide treatment also enabled the introduction of additional nitrogen-containing functional groups to the biochar structure, leading to enriched elemental nitrogen content (Hafeez et al. 2022). Similarly, salts (e.g., chlorides and phosphates) and other substances (e.g., hydrogen peroxide) have also been studied to modify textural parameters and surface chemistry of biochar. Salt treatment can affect the biochar surface area and pore size, leading to the formation of new active sites that contain metals on the biochar surface. Liu et al. impregnated the pine-based biochar with $ZnCl_2$ and reported an increase in BET (by ~ 174%) and TPV (by ~ 94%), while the APS was observed to decrease by ~ 32%. The use of salts in the biochar modification can also result in reduced textural parameters. Breton et al. reported that the modification of Eucalyptus tree biochar using magnesium chloride decreased biochar BET from ~ 1.49 to ~ 0.97 m^2 $g^{-1}$and TPV from ~ 0.43 to ~ 0.30 cm^3 g^{-1} (Arbelaez Breton et al. 2021). Wang et al. reported that chlorides clogged the pores when attached to the biochar surface after treatment (Wang et al. 2015). Hydrogen peroxide treatment is another method for biochar property modification. Tan et al. reported a decreased BET surface area and increased TPV and APS in biochar after treatment with H_2O_2 (Tan et al. 2019). H_2O_2 treatment led to a higher elemental O/C ratio of biochar because H_2O_2 generates additional carboxylic, lactone, and hydroxyl groups (Huang et al. 2016; Huff and Lee 2016). H_2O_2 has the advantages of being environmentally safer and less expensive than strong acids, bases, and salts.

Chemical modifications of biochar hold many opportunities for tailoring the biochar to a particular applications. Oftentimes, biochar is utilized as an environmentally conscious and renewable resource in ecological technologies. In such cases, polymers are typically hydrophilic in nature due to being biodegradable or compostable. Thus, chemically treated biochar with increased oxygen functional groups will enhance its compatibility with hydrophilic polymers. Many traditional polymers, however, are often hydrophobic and require a hydrophobic biochar for better compatibility.

These differences further emphasize the need to tailor biochar to specific polymers through chemical treatments for 3D printing applications.

5 Summary and outlook

In this study, the emerging area of biochar/polymer 3D-printed composites is reviewed. The mechanical and thermal properties of the resulting composites and any challenges that occur due to biochar incorporation were highlighted. In particular, biochar composites worked well mechanically at limited biochar concentrations (< 10 wt.%) but aggregated and failed under larger concentrations. This can complicate composite formulations because, to act as a filler, biochar will need to replace large portions (> 10 wt.%) of the matrix for a more significant impact on polymer reduction. In other words, low concentrations have the advantage of improved properties, but high concentrations have the advantage of a bio-based and renewable filler. As an additive, these small concentrations may be sufficient to produce adequate properties but this depends heavily on applications. Aggregation also faced potential occurrences within the filaments and during the extrusion process. This often leads to nozzle clogging and interruptions of the printing process. On a more positive note, rheological studies have shown that biochar improves polymer flow from the nozzle. In general, 3D printing also faces difficulties with layer adhesion, and while some progress has been made regarding improvement due to biochar incorporation, conflicting reports of its benefit to this aspect remain. Biochar-based 3D printed polymer composites hold potential for many applications (including packaging, automotive, aerospace, and construction) and therefore require further investigation into potential ways to improve their processability.

The scope of this work included the analysis of current 3D biochar/polymer composites, with a significant focus on biochar production methods for improved 3D printing in accordance with the problems discussed above. For pyrolysis to produce the biochar, it was determined that lower temperatures could produce a greater yield, whereas higher temperatures produced a greater surface area for interactions with the polymer. Feedstock composition can vary the biochar makeup, which can affect its interaction with the polymer and should be carefully considered. For post-production modifications, biochar can be functionalized using mechanical or chemical methods. The former has largely focused on ball milling; a process well-suited for the laboratory environment. Ball milling not only reduces particle size for a more homogenous material with less aggregation but also tailors functional groups for a particular polymer compatibility. Furthermore, chemical modifications can adjust biochar surface area while modifying surface functional groups for chemical interactions.

Regarding 3D printing of biochar/polymer composites as well as biochar production and functionalization methods, this review presented a set of recommendations for future investigations as well as what still needs to be investigated in this emerging area. For biochar production, lower temperatures may be best for a high yield with lower energy input, with greater surface area achieved by mechanical and chemical methods in post-processing. The feedstock, produced from waste material, is recommended to be woody biomass such as that from forestry industries. The high lignin content can produce a higher surface area as well as limit ash content to reduce aggregation potential and, ultimately, nozzle clogging. Ball milling offers a simple processing method for reducing biochar particle size (and, therefore, improving dispersion) and simultaneously increasing surface area. Polymer compatibility plays a significant role in processability, with many of the currently investigated polymers being hydrophobic (for example, PP, PET, HDPE, PLA, etc.). Because of this, ball milling is recommended to be done in an atmosphere without oxygen (such as in a vacuum or nitrogen), so the surface is limited in oxygen moieties. For polymers that are more hydrophilic, ball milling in an oxygen-containing atmosphere may be beneficial, as well as chemical treatments with acids (e.g., HCl, H_2SO_4, H_3PO_4, HNO_3, etc.). This may help to functionalize the biochar to be more compatible with hydrophilic polymers.

Presently, there is limited research into 3D-printed biochar/polymer composites, and future research should focus on several aspects of its investigation. In particular, varying the production methods of biochar (pyrolysis conditions and feedstock/feedstock composition) and determining their effects on composites needs further investigation. There is little information on this for 3D-printed biochar composites and for the category as a whole, regardless of polymer processing techniques. Such research needs to examine the effects of changing pyrolysis parameters as well as feedstock type in order to directly compare their effects on the composites. Similarly, while there is much research on biochar mechanical and chemical modifications, there are few direct comparisons where 3D-printed parts are examined according to varying modification methods. This is important as it can potentially affect the processability of the printed specimens. Due to this being an emerging area, there is much research that still needs to be done, where biochar characteristics can be examined and linked to 3D printing processability.

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