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ADDITIVE MANUFACTURING OF WHEAT STRAW FOR SUSTAINABLE THERMAL INSULATION APPLICATION

Zipeng Guo
Industrial and Systems Engineering
Rochester Institute of Technology
Rochester, NY
zxgeie@rit.edu

Licheng Liang
Industrial and Systems Engineering
State University of New York at Buffalo
Buffalo, NY
lichengl@buffalo.edu

Jason Armstrong
Mechanical and Aerospace Engineering
State University of New York at Buffalo
Buffalo, NY
jna4@buffalo.edu

Shenqiang Ren
Materials Science and Engineering
University of Maryland
College Park, MD
sren@umd.edu

Chi Zhou
Industrial and Systems Engineering
Cell, Gene and Tissue Engineering Center
State University of New York at Buffalo
Buffalo, NY
chizhou@buffalo.edu

ABSTRACT

Thermal insulation materials reduce heat transfer and are typically made from materials like fiberglass, foam, or mineral wool, which are engineered to trap air and hinder heat conduction and convection. The traditional manufacturing processes of thermal insulation materials are often energy-intensive and result in significant greenhouse gas emissions. In the current global drive for sustainability, these energy-intensive manufacturing processes raise environmental concerns and need to be addressed. In this work, with the objective of addressing both material sustainability and manufacturing sustainability, we present an additive manufacturing strategy to fabricate biomass materials for thermal insulation applications. Firstly, we propose to use biomass materials, such as wheat straw, as the primary feedstock materials for manufacturing. Such biomass materials offer the unique capacity to sequester carbon dioxide during their growth, and when incorporated into thermal insulation structures, they effectively capture and store carbon inside the structure. Concurrently, our pursuit of manufacturing process sustainability is driven by using a cost-effective additive manufacturing technology to fabricate durable thermal insulation structures. In the presented work, we first demonstrate the formulation of a 3D-printable ink using chopped straw fibers. We conduct comprehensive rheological characterizations to reveal the shear-thinning properties and the printability of the straw fiber ink. Utilizing the direct ink writing (DIW) process, the straw fiber material is deposited into 3D structures. Following bulk material characterization tests, including microstructure, mechanical, and thermal tests. We unveil the low thermal conductivity and robust mechanical properties. This paper marks the first work of 3D printing of wheat straw fibers for thermal insulation structures. The discoveries in this pilot work demonstrate the potential to leverage additive manufacturing technologies and sustainable biomass materials to create both functional and value-added wheat straw parts tailored for thermal insulation applications.

Keywords: Additive Manufacturing; Biomass Materials; Sustainable Manufacturing; Direct Ink Writing; Non-Newtonian Shear-Thinning Property; Thermal Insulation

NOMENCLATURE

G'	Storage Modulus
G"	Loss Modulus
q	Heat flux
K	Thermal conductivity

1. INTRODUCTION

Thermal insulation is vital in various industries as it mitigates the inherent nature of thermal diffusion and minimizes the heat transferring from a higher temperature environment to a lower temperature environment. Using thermal insulation materials could conserve energy by maintaining thermal equilibrium and preventing excessive heat loss or gain [1]. Energy conservation not only leads to economic savings but also translates to reducing greenhouse gas (GHG) emissions and lowering carbon footprints [2, 3]. The global thermal insulation market, valued at approximately $31.4 billion in 2022, is projected to reach $96.0 billion by 2028 [4, 5]. This rapid growth is driven by increasing awareness of energy efficiency and sustainability. Among the various thermal insulation materials, fiberglass, mineral wool, and synthetic polystyrene boards are widely used [6, 7]. The manufacturing processes for these insulation materials are well-established, where melting, extrusion, or chemical reactions are used to create the insulation products [8]. For instance, fiberglass insulation is produced by melting glass and then spinning it into fibers. Polystyrene boards are made by melting and steam-expanding polystyrene beads [9]. However, it is important to note that these traditional manufacturing processes for insulation materials typically demand significant energy consumption and rely on fossil oil-based resources. For example, extracting raw materials, such as sand for glass or petroleum for plastics, involves energy-intensive processes. Melting these raw materials to create insulation products demands substantial thermal energy obtained from fossil fuel sources. These manufacturing practices result in greenhouse gases emissions into the atmosphere and have negative effects on the environment and climate change. In the context of today's global emphasis on sustainability and carbon neutrality, it becomes imperative to optimize these processes by implementing energy-efficient technologies and promoting recycling initiatives [10]. This seeming paradox, wherein energy-intensive processes are employed to fabricate energy-conservation materials, warrants critical consideration.

However, transitioning to sustainable manufacturing practices faces two major challenges: material sustainability and manufacturing sustainability. (1) Material sustainability challenge: Insulation materials often rely on non-renewable resources such as petroleum-based plastics or energy-intensive raw materials like sand for glass [11-13]. Ensuring a sustainable supply chain for these materials while minimizing their environmental impact is a major challenge. Moreover, developing recyclable or biodegradable alternatives introduces additional material sustainability challenges. (2) Manufacturing sustainability: The reduction of energy consumption in the manufacturing of insulation materials poses significant challenges. Energy-intensive manufacturing processes release greenhouse gases and other pollutants into the atmosphere. Developing more efficient manufacturing methods to reduce

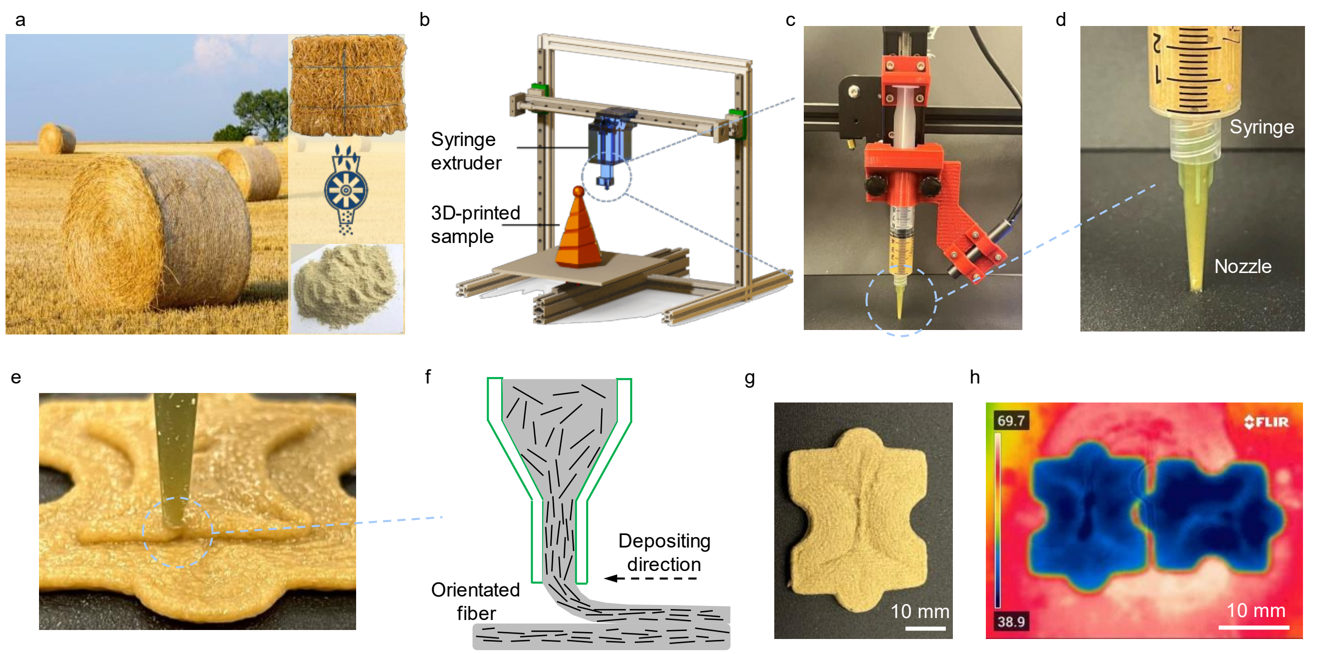

Figure 1 | Additive manufacturing process of wheat straw fiber for thermal insulation structures. (a) Utilizing raw wheat straw as feedstock materials. The fine fibers are produced through hammer milling. (b) Schematic representation of the Direct Ink Writing (DIW) process. (c) A photograph of the syringe extruder. (d) Close-up view of the syringe-nozzle configuration. (e) Photograph showing the DIW printing process. (f) Schematic illustration of the fiber alignment during extrusion printing. (g) A 3D-printed straw fiber puzzle sample. (h) Infrared (IR) image showcasing the thermal insulation performance of the assembled puzzle pieces.

emissions is crucial for sustainability [13]. The research of Shuang [14] et al. showed that the main components of wheat straw lignocellulose are cellulose, hemicellulose, and lignin. The non-disordered structure of cellulose indicates that it is biodegradable. The microbial pretreatment method has the advantages of low energy consumption and fewer chemical additives. Microorganisms including fungi have good biodegradability for the removal of lignin in wheat straw materials.

In an effort to address sustainability challenges, significant attention is drawn to eco-friendly materials due to their minimal negative impact on the environment during production, use, and disposal. Eco-friendly materials are often derived from renewable resources, such as plants or recycled materials. Among various eco-friendly resources, biomass materials have gained considerable attention as an alternative to conventional fossil fuel-based materials. Biomass materials are derived from plants and biological sources. Unlike finite resources like fossil fuels, which take millions of years to form, biomass materials, such as wheat straw, bamboo, and agricultural residues, can be harvested and replanted regularly, thus providing sustainable and renewable resources [15]. Biomass materials often possess fibrous structures. Researchers have discovered that the inherent fibrous structures in materials like wheat straw, bamboo, and wood are arranged in a porous morphology, creating air pockets within the biomass materials. These air pockets restrict the movement of heat and reduce the thermal conductivity of the material. As a result, biomass-based thermal insulation materials are highly sought after for applications in thermal insulation. Rojas et al. developed thermal insulation structures using wheat straw and corn husk fibers [16]. The process involved pulping the fibers into a slurry and drying them in molds. It was found that cellulose, hemicellulose, and lignin are major constituents in these biomass materials, wherein lignin forms rigid fibrous structures. They have also discovered that fiber length had a significant impact on the thermal conductivity of the materials, with shorter fibers leading to decreased thermal conductivity. The microstructures of wheat straw were investigated by Yin et al. [17]. Hierarchical porous structures were observed in raw wheat straw with the cellular diameter of wheat straw progressively reducing from the core towards the external surface (epidermis). Lignin and hemicellulose were found to be the binding agents that held the cells together. These hierarchical porous structures were identified as key contributors to the low thermal conductivity of raw wheat straws. Although the conductivity of biomass-based insulation materials is comparable with conventional insulation products, they face issues such as flammability and moisture absorption [18]. To this end, research efforts are invested in developing biomass-based composites. Cen et al. investigate a full bio-based lignin composite aerogel for thermal insulation and flame retardancy [19]. With abundant carbon in lignin, when exposed to high temperatures, carbon-containing materials tend to char and form a protective barrier that inhibits the spread of flames, thus showing flame retardancy. Moreover, the rigid nature of lignin can effectively reinforce mechanical strength. Wang et al. developed synthesized lignin aerogel, and discovered that the porous microstructures reinforced by lignin exhibit robust mechanical properties, thermal insulation properties, and sound absorption properties [20]. Moisture adsorption is a potential issue with biomass-based thermal insulation structures, as water moisture could reduce the thermal insulation performance and affect the durability of the structural strength. An effective solution is using hydrophobic surface modification. Chemical vapor deposition of methyl trichlorosilane (MTCS) [19], hexamethyldisilazane (HMDS) [21], and carnauba wax (CW) and beeswax (BW) [22] are widely used in research.

Conventional manufacturing processes, such as molding, laminating, extrusion, and foaming, have been employed to fabricate biomass materials. However, these methods have long relied on energy-intensive procedures, such as high-temperature processing, extensive drying and curing, and high-pressure compression. These energy-intensive practices were established well before the urgency of climate change became a global concern. Recently, the concept of a circular economy (CE) has emerged as a model to address sustainability in manufacturing [23-25]. The circular economy model advocates for resource optimization, waste reduction, and promotes sustainable practices throughout the lifecycle of products [26]. It aligns with the growing need for environmentally responsible manufacturing methods. One notable challenge that limits conventional manufacturing techniques from adopting the CE model is their centralized nature, often leading to significant supply chain and transportation costs [27-29]. In this regard, additive manufacturing emerges as a decentralized solution towards a cost-effective circular economy. Additive Manufacturing (AM), also known as 3D printing, is an advanced manufacturing process that constructs three-dimensional objects layer by layer. By additively fabricating 3D structures directly from digital files and only depositing materials as needed, AM can largely reduce material waste and enable on-demand production. Moreover, AM allows for decentralized manufacturing [29-31]. Instead of relying on a centralized factory, products can be manufactured closer to the end-users, reducing the need for long-distance shipping and its associated environmental impact. However, employing AM technologies to fabricate biomass materials poses challenges. Many AM technologies are initially developed for synthetic polymers or metals and may not be inherently suited for natural biomass materials. Research advances have focused on using polymer-biomass composite materials for AM feedstocks. Bi et al. summarized the methods of 3D printing natural fiber composite materials and their applications in biomaterials [32]. Composite materials are made by adding natural fibers to Polylactic acid (PLA). And it can be printed using the fused filament fabrication (FFF) process. Oksman et al. mixed flax fiber with PLA to produce environmentally friendly fiber/polypropylene filament [33]. Another method is the 3D printing of fiber-based composite hydrogel materials. Kajsa et al. formulated a bio ink that combines the excellent shear-thinning properties of nano fibrillated cellulose (NFC) with the properties of alginate materials [34]. The combination of rapid cross-linking capabilities was used for 3D bioprinting of living soft tissues and cells. While these composite structures exhibit robust mechanical strength, the addition of polymeric materials leads to higher thermal conductivity and carbon footprint, limiting its functionality in thermal insulation and impact on sustainability.

In this work, our objective is to address both materials sustainability and manufacturing sustainability for thermal insulation materials. We use straw fibers as the feedstock material, and extrusion-based direct ink writing process to fabricate thermal insulation structures. The straw fibers are derived from the residual stalks of wheat plants after the grains have been harvested. In many agricultural practices, straw is often left in the field or disposed of as agricultural waste. By repurposing the wheat straw, we can reduce agricultural waste and 3D printing eco-friendly thermal insulation materials. We design the printable straw fiber ink and utilize the direct ink writing (DIW) process to fabricate the 3D structures. DIW is a facile 3D printing technology to deposit viscous inks to 3D geometries [35, 36]. It allows for precise material deposition, reducing waste and enabling on-demand production. In order to assess the printability of the straw fiber ink, we perform rheological characterization to reveal the shear-thinning properties and the viscoelastic properties of the ink. In addition, we performed thermal and mechanical properties characterization on the 3D-printed straw fiber samples. The results demonstrate that the wheat straw-based insulation structure exhibits a low thermal conductivity of 0.030 W/m K and a robust mechanical strength of 15.89 MPa. The methods to fabricate the insulation straw are described in this paper, and a comprehensive investigation and discussion of the sample properties are presented. We also share our perspectives on the outlook of additive manufacturing of straw-based thermal insulation. Notably, this paper marks the first work of 3D printing of wheat straw fibers for thermal insulation application with promising potential to scale up for building insulation envelopes. The promising discoveries in this pilot work demonstrate the potential to leverage advanced manufacturing technologies and sustainable biomass materials to create both functional and value-added wheat straw parts tailored for thermal insulation applications.

2. MATERIALS AND METHODS

2.1 Straw fiber ink formulation

First, the raw wheat straw (EZ-straw, Rhino Seed) was hammer-milled into straw fiber and then filtered with a sieve (50 mesh) to remove excessively long fibers which will block off the nozzle during printing. The processed straw fiber was added to deionized water at a ratio of 17wt%, and then 1.5 mL of dispersing agent (DARVAN 811; Vanderbilt Minerals) was added and stirred for 3 hours until a uniform slurry was obtained. Finally, 3g of cellulose nanofiber (92% Crystallinity; Nanografi) was added into the solution in three batches and stirred evenly to obtain a printable straw fiber ink. Alternatively, fumed silica can be used as an alternative to cellulose to increase viscosity to facilitate the extrusion-based 3D printing process.

2.2 Rheological characterization

The rheological properties of the straw fiber inks investigated in this manuscript were characterized at room temperature with a TA HR30 rheometer (Waters Corporation, USA), using a 20 mm parallel plate geometry set at a gap of 250 µm. Flow rate sweep tests were carried out with a shear rate from 10^{-1} to 10^{2} s^{-1}. Viscoelastic behavior was evaluated by oscillatory measurements at 1 Hz and with a strain range from 0.01% to 1000%.

2.3 Direct ink writing process

A fused filament fabrication (FFF) printer (Ender 3 Pro, Creality 3D) is retrofitted to the direct ink writing process. The original heating-extruder assembly was replaced with a motorized linear stage (NEMA 11) and a custom-designed fixture for syringes (10mL luer lock, LiteTouch). The motorized linear stage was used to move the piston of the syringe, generating a downward extrusion force for extrusion by pushing the ink through the nozzle. A luer lock nozzle (1.54 mm diameter, Nordson EFD) was attached to the syringe. The straw fiber ink was loaded into the syringe and mounted onto the fixture. The motion of the syringe was guided by G-code files generated using an open-source software Slic3r. The thickness of each layer is 0.85 mm. Following printing, the as-printed samples could be dried in ambient environments under room temperature or dried using a freeze-drier with pressure maintained at 0.013–0.018 mBar.

2.4 Mechanical characterization

The uniaxial compression tests were conducted using a universal test system (Model SSTM-20KN from United Testing Systems).

2.5 Thermogravimetric analysis (TGA)

The thermogravimetric analysis (TGA) was carried out using a TA Instruments SDT Q600 Differential Calorimeter/Thermogravimetric Analyzer (DSC/TGA) under airflow. The temperature increased from room temperature to 500 °C at a ramp rate of 20 °C/min.

2.6 Thermal conductivity measurement

The thermal conductivity of the straw-fiber samples was measured using a custom-built instrument with heat flux sensors (FluxTeq). The heat flux and thermal conductivity relationship was given by:

$$q = \frac{K\Delta T}{\Delta x} \quad (1)$$

K was the thermal conductivity of the sample. The sample was placed in between two parallel plates, the temperature difference of the plates was ΔT. Δx was the sample thickness. q was the heat flux value.

2.7 Microstructure characterization

The microstructures of the printed straw fiber specimen were imaged using the Carl Zeiss AURIGA scanning electron microscopy (SEM). A thin layer of gold was sputter-coated on the surface of interest to avoid the charging effect during SEM imaging.

3. RESULTS AND DISCUSSION

3.1 Rheological property and printability

The shear rate sweep tests were carried out with two types of straw fiber inks, including ink of straw fiber with cellulose and ink of straw fiber with fumed silica (Fig. 2 (b)). In this test, ink viscosities were measured as a function of shear rate, and the results were plotted in Fig. 2. The log-log scale plots in Figs. 2(b) and (c) illustrate a linear decreasing trend, indicating that as shear rate increases, viscosity decreases due to the restructuring of the fluid structure—a phenomenon known as shear-thinning. When no shear force is applied, the 2D-like straw fibers align randomly, contributing to a higher viscosity due to increased interactions between the fibers [37]. However, as shear force is introduced, the 2D-like fibers start to reorient or disperse in the direction of the shear, leading to a decrease in viscosity (Fig. 1(f)). This reorientation, combined with the cellulose and fumed silica network, contributes to the observed shear-thinning behavior. In Fig. 2(c), the comparisons highlight that increased cellulose concentration corresponds to higher viscosity. Both cellulose and fumed silica were introduced into the ink to tune its rheological properties. Cellulose is a carbohydrate material that forms a continuous network when dissolving into the ink. This network creates more entanglements between cellulose molecules, acting as thickening agents to hinder the movement of ink flow and causing an increase in viscosity. This effect is more pronounced at higher cellulose concentrations, where the formation of a more extensive network leads to greater viscosity (Fig. 2(c)).

On the other hand, the dynamic rheological behavior is shown in Figs. 2 (d)-(f), where the Storage Modulus (G') measures the ink's ability to store energy, and it represents the elastic (solid-like) behavior of the ink. The Loss Modulus (G") measures the ink's ability to dissipate energy, and it represents the viscoelastic (liquid-like) behavior of the ink. From the plots, both the G' and G" remain constant under low strain (<1%), indicating the ink behaves like solid and remains structural integrity. As the oscillation strain further increases, ink viscosity decreases, leading to decreasing Storage and Loss Moduli. The crossover points between the G' and the G" are the yield points, indicating that the inks are transitioning from solid gel yield to liquid flow. The yield point is used to characterize the viscoelastic property of the ink. As the cellulose concentration increases, as shown in Figs. 2(d) to (f), the yield points shift from 9.1% strain to 10.5% strain, and further to 50.4% strain, indicating that increasing cellulose leads to a more pronounced solid-to-liquid transition.

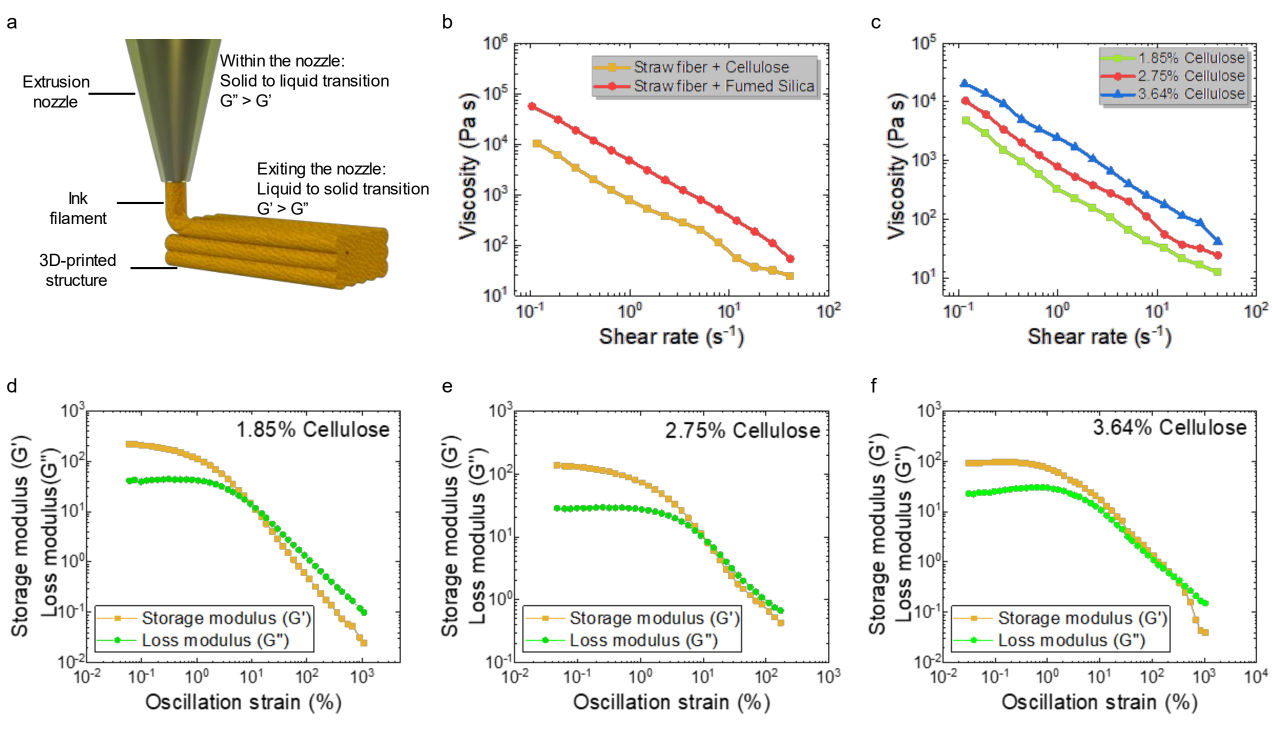

Figure 2 | Rheological characterization of the straw fiber inks. (a) A schematic illustration of the relationship between the ink extrusion and the rheological properties. (b) Shear thinning properties of straw fiber inks with cellulose and fumed silica as different rheological modifiers. (c) Shear thinning properties of the straw fiber inks with varying concentrations of cellulose. (d)-(f) Dynamic rheological characterization of the straw fiber inks with varying concentrations of cellulose.

The rheological analysis indicates that utilizing cellulose as a rheological modifier in the range of 1.85% to 3.64% is optimal for facilitating the extrusion-based DIW process. Below 1.85% cellulose concentration, the ink exhibits insufficient viscosity and storage moduli to support forming a robust 3D structure. Conversely, exceeding 3.64% cellulose concentration results in an ink that is overly thick for stable and continuous filament extrusion. In Section 3.4, we will discuss how varying cellulose concentrations influence the properties of the samples.

Reproducibility and repeatability are important considerations in producing a transformative new manufacturing process. To demonstrate reproducibility and repeatability, we printed printing tests at independent times on different days, generating a total of seven printed samples. The random variation in environmental conditions, such as the humidity and temperature, was considered independent variables. Our goal is to have consistent printing results and qualities under diverse environmental conditions. Upon fabrication, we meticulously measured the length, width, height, and weight of each sample, presented in Fig. 3. Consistent results can be observed in the histogram plots in Fig. 3 to demonstrate the reproducibility and repeatability. There are usually two reasons why a print is not repeatable. Firstly, nozzle blockage will prevent printing. We use mesh to filter out the excessively long wheat straw fiber. Secondly, air bubbles mixed in during the loading process will also affect the printing effect. Our treatment method is to process the syringe filled with materials at low pressure for 5 minutes to reduce air bubbles in the syringe.

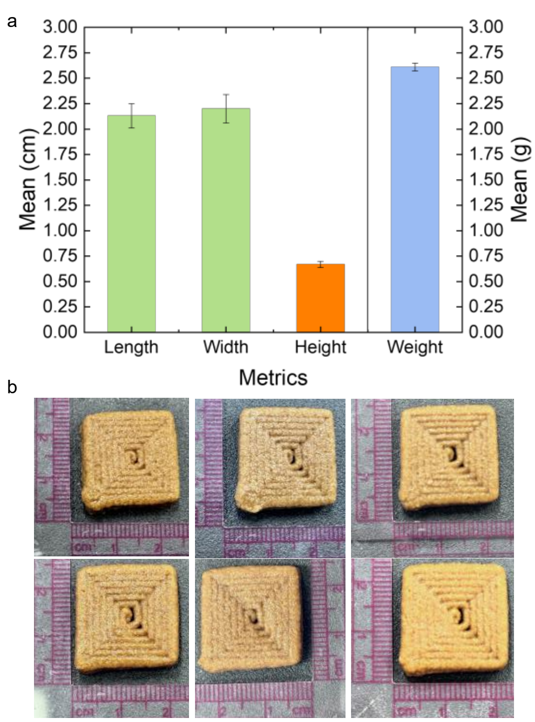

Figure 3 | Reproducibility study of the straw fiber additive manufacturing process. (a) A summary of the mean and standard error of the mean for length, width, height, and weight of the 3D-printed straw fiber samples. (b) Photographs of the printed samples in this study.

3.2 Direct ink writing (DIW) process

We printed various 3D geometries, as presented in Fig. 4, to highlight the versatility of the DIW process with straw fiber inks. The hollow cylinder model in Fig. 4(b) demonstrated that a high aspect ratio (height-to-wall thickness ratio) structure can be printed and maintained freestanding. This model highlights that the straw fiber inks have sufficiently high storage modulus (G') to sustain a 3D structure without sagging or drooping. While the visibility of each layer warrants future process resolution optimization, it's essential to emphasize that achieving high aspect ratio structures demands precision in the DIW process. Any deviation from the defined path can become more pronounced with taller structures, potentially leading to misalignment and compromising print quality [35, 38]. Figure 4(b) serves as an example of a printed sample.

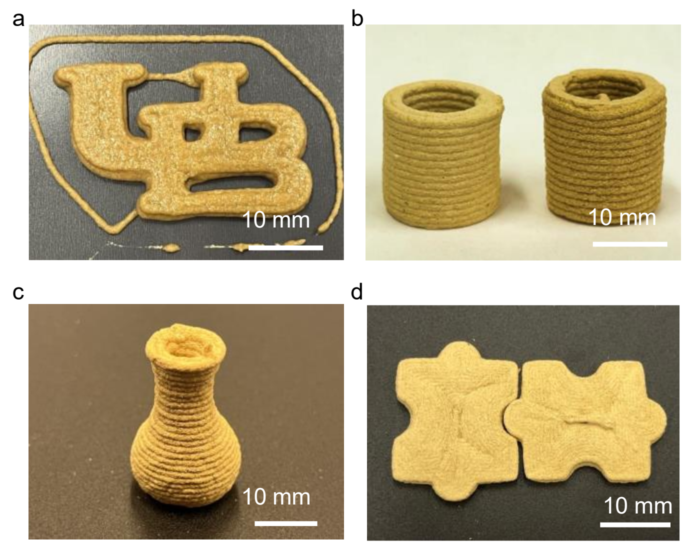

Figure 4 | 3D-printed straw fiber structures. (a) University at Buffalo (UB) logo. (b) Hollow cylinder model. (c) Vase model. (d) Assembled puzzle pieces.

The 3D-printed vase model in Fig. 4(c) has varying geometry along the vertical direction (Z-axis). This sample demonstrates that even with overhanging structures, the straw fiber ink can uphold freestanding structures without requiring additional supporting structures. Overhanging structures, often referred to as downfacing structures, pose challenges in DIW printing and usually need support structures to prevent deformation or collapse during printing. Support structures will be removed in post-processing and essentially create material waste. However, owing to the high storage moduli, the straw fiber inks exhibit a remarkable ability to self-support and retain their shape without the necessity of additional support structures.

We printed and assembled puzzle pieces as shown in Figs. 1(g) and 4(d) to demonstrate the potential for modularization and self-assembly in DIW-printed structures. The modular nature of the puzzle-like structures and its interlocking mechanism opens avenues for constructing larger structures, such as building insulation walls, without the need for fabricating large pieces and transporting large products. Moreover, by standardizing these modular components, it becomes possible to create a collection of building blocks that can be assembled into diverse structures.

3.3 Microstructure characterization

Scanning electron microscopy (SEM) was employed in this test with two goals: firstly, to reveal the microstructure of the 3D-printed straw fibers samples, and secondly, to discover the structural distinctions between the freeze-dried (FD) samples and the ambient pressure dried (APD) samples. The FD samples exhibit a porous morphology (Fig. 5(a)-(b)). This is due to the freeze-drying process conducted in a low-pressure, low-temperature environment, enabling the sublimation of ice directly into gas and bypassing the liquid phase [37, 39]. The porous structures observed in the FD samples originate from the removal of ice during the freeze-drying process [40, 41]. The ice sublimation results in low surface tension and leads to minimal shrinkage of the samples.

In contrast, the APD samples (Fig. 5(c)-(d)) demonstrate a more condensed morphology with a noticeable volume shrinkage of 40%. During APT, the higher pressure increases the rate of water evaporation, causing water molecules to escape from the liquid more rapidly than in FD. The increased presence of surface tension in APD may intensify shrinkage, as surface tension tends to pull liquid molecules together, forming cohesive forces. As the liquid evaporates, this cohesive force contributes to shrinkage and densification of the straw fiber samples.

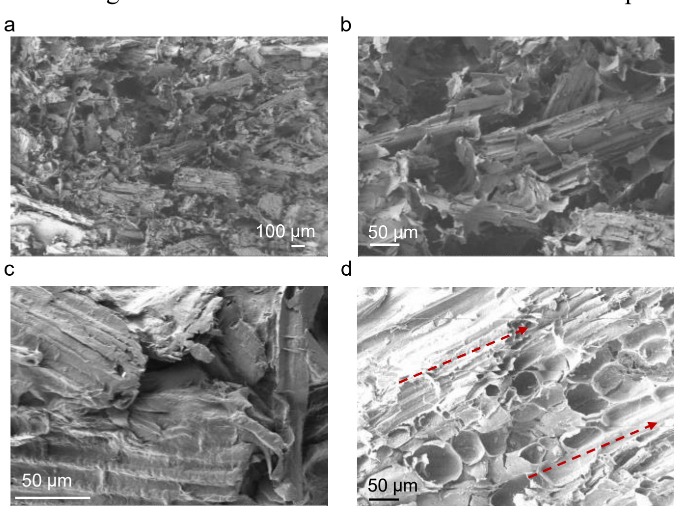

Figure 5 | Microstructural characterization of the 3D-printed straw fiber structures. (a) and (b), Morphology of the freeze-dried samples. (c) and (d), Morphology of the ambient pressure dried samples.

From SEM images in Fig. 5, we observe the straw fibers have aligned patterns along the extrusion direction. As the ink is extruded, the shear rate and shear forces are generated. The shear forces act parallel to the direction of the straw fiber ink flow, thus orientating and aligning the fibers along the direction of the shear, resulting in the observed uniformity in the microstructure [42]. Aligned fibers could contribute to enhancement in structural integrity with increased strength and resistance along the direction of alignment. This alignment also influences the anisotropic properties, making it more robust and tailored when subject to external loads in the same direction.

3.4 Property characterization

The properties of 3D-printed straw fiber samples were comprehensively investigated through thermogravimetric analysis (TGA), compressive mechanical testing, and thermal conductivity measurements. The TGA test shown in Fig. 6(a) revealed that the straw fiber samples have several stages of thermal decomposition. The initial stage from room temperature to 100 °C has a 6% weight loss attributed to moisture removal. Then from 100°C to 250°C, the weight remains close to constant, indicating a stable behavior within this temperature range. The first two stages imply the potential practical application of straw fiber samples, particularly in thermal insulation, owing to their robustness in the temperature spectrum. Upon further heating from 250°C to 300°C, a rapid weight loss was observed, primarily attributed to the decomposition of cellulose component from both the raw wheat straw and additives, as well as hemicellulose component from the wheat straw. Then future weight loss in higher temperatures was a result of lignin decomposition. The remaining 15% manifested as chars, which is the carbonaceous residue left after the volatile components have been released. We used the TGA test to discover the thermal decomposition stages and also provide proof that the 3D-printed samples consisted of organic ingredients. Our approach involves the exclusive use of organic substances without the incorporation of inorganic insulation materials such as silica and fiberglass. This material composition ensures an eco-friendly production process, avoiding energy-intensive manufacturing. Additionally, the organic nature of the substances used allows for easy disposal at the end of the product's life cycle.

Compressive mechanical tests were conducted to evaluate the structural integrity of the 3D-printed straw fiber samples. As shown in Fig. 6 (b) and (c), a comparative analysis was performed between the freeze-dried (FD) and ambient pressure dried (APD) samples. Additionally, we examined the influence of cellulose concentration on the mechanical properties of each drying method. From the comparison, we found that the APD samples show higher stiffness and strength. The maximum Young's modulus was 15.89 MPa with a compressive strength of 7.87 MPa. Moreover, an increase in cellulose concentration correlated with enhanced Young's modulus and compressive strength. Cellulose molecules are self-assembled by strong intermolecular forces [43-45]. When cellulose concentration increases, there are more intermolecular interactions, creating a tighter and more cohesive structure. Besides, cellulose may enhance the adhesion between layers, promoting better layer-to-layer bonding [37, 46]. Together, these two factors lead to an overall increase in mechanical properties.

In contrast, the FD samples have comparatively lower mechanical properties, primarily due to the freeze-drying process inducing a more porous structure (Fig. 5(a)). The porous structures contain lots of void space, thus compromising overall mechanical properties compared to the more compact and structurally robust APD samples.

The thermal conductivity tests were carried out to assess the thermal properties of the 3D-printed straw fiber samples. As shown in Fig. 6(d), the lowest thermal conductivity reached 0.030 W/mK. When compared to commercially available thermal insulation products, the 3D-printed straw fiber specimens show a lower thermal conductivity, indicating superior insulation performance [35, 38, 47-49]. In the case of freeze-dried (FD) samples, as the concentration of cellulose increases, the thermal conductivity remains at the same level, as the porous structures of the FD sample dominate the thermal properties. In porous structures, heat transfer can involve the movement of air within the pores. Air convection in an ambient environment is a very low process, thus contributing to low thermal conductivity. In contrast, for the APD samples, the addition of cellulose significantly influences the thermal properties. This is primarily due to cellulose forming a more condensed structure, essentially enhancing the solid conduction of heat [50, 51].

Through a comprehensive analysis of both mechanical and thermal characteristics, it becomes evident that achieving an optimal balance in cellulose concentration is crucial for 3D-printing thermal insulation materials. The concentration of cellulose emerges as a key factor, impacting not only the printability but also the mechanical and thermal properties of the straw fiber specimens. Higher cellulose concentrations enhance mechanical strength, contributing to improved structural integrity. However, this strength enhancement comes at the expense of thermal insulation performance, where higher cellulose concentrations tend to compromise the thermal insulation properties.

Future improvements could be focused on finding a cellulose concentration that strikes an equilibrium, ensuring favorable printability, robust mechanical strength, and thermal insulation performance. Another crucial aspect for future exploration is the assessment of the carbon footprint entailed in the life cycle of the insulation straw fiber samples. A Life Cycle Assessment (LCA) will serve to quantify the environmental impact across multiple stages, including wheat straw fiber processing, additive manufacturing, energy consumption in manufacturing, transportation, energy savings with the insulation materials, and the end-of-life disposal process.

3.5 Perspective and Outlook

This pilot work showcases the viability and efficiency of employing additive manufacturing to produce thermal insulation structures with wheat straw fibers. Beyond its immediate application, the use of wheat straw as a representative

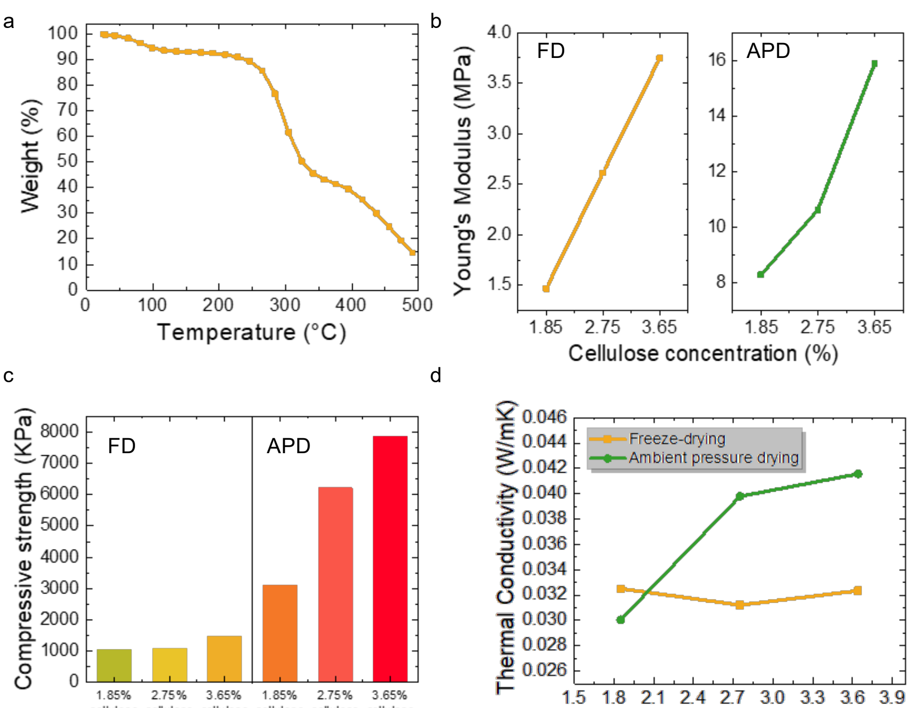

Figure 6 | Sample properties characterization. (a) Thermogravimetric analysis (TGA) of the 3D-printed straw fiber samples. (b) Young's modulus of the straw fiber samples with varying cellulose concentrations. (c) Compressive strength of the straw fiber samples with varying cellulose concentrations. (d) Thermal conductivity of the 3D-printed straw fiber structures.

of biomass materials presents an emerging opportunity to use sustainable material to fabricate functional structures. The future development of AM biomass materials will focus on large-scale, integrated AM systems to fabricate value-added biomass materials. In the sections below, we share our perspectives and outlooks for future advancements:

(1) **Large-scale fabrication**: Transitioning from lab-scale prototypes to large-scale production is a pivotal advancement in the application of biomass structures. Large-scale fabrication leads to tangible structures applicable in construction, packaging, and diverse industries. For instance, biomass materials with low thermal conductivity can be integrated into building envelopes positioned between exterior and interior walls. They serve as insulation barriers to improve energy efficiency and promote sustainable construction practices.

(2) **Decentralized manufacturing:** Additive manufacturing features the novel idea of decentralized manufacturing [31, 52]. This approach facilitates the production of biomass structures at local levels, potentially on the site where raw materials are harvested. The shift away from centralized factories reduces transportation costs and the associated environmental impacts, but also potentially fosters local economies. This decentralized model offers increased flexibility in responding to regional demands, paving the way for a more adaptive and sustainable manufacturing ecosystem. However, achieving decentralized manufacturing would take a long path ahead. It's important to note that while AM offers significant advantages in terms of decentralization, it might not replace traditional manufacturing entirely. Instead, it complements existing methods and is particularly useful in situations where customization, rapid production, or localized manufacturing is essential.

(3) **Integrated systems:** By integrating various stages in the AM process, including material processing, 3D Printing, and post-processing, an integrated system ensures quality control of the insulation straw fiber structures [53]. It streamlines the production chain, from sourcing raw materials to delivering biomass-based products, thus offering the foundation for a robust and sustainable manufacturing system.

(4) **Life cycle assessment (LCA):** Incorporating Life Cycle Assessment (LCA) into AM for biomass materials is a proactive step toward sustainability [8]. Understanding and mitigating the environmental impact at every stage of production aligns with the growing emphasis on sustainable practices. LCA could guide environmentally responsible manufacturing and also inform consumers and industries about the ecological credentials of biomass-based products. Additionally, coupling LCA with the carbon emissions that are associated with material processing, manufacturing, energy consumption, transportation, and end-of-life disposal. It offers a transparent disclosure of the ecological footprint of biomass-based products. It’s worth noting that the straw fiber acts as a carbon sink through photosynthesis during its growth, and its adoption in insulation materials sequesters carbon and stores carbon within buildings. Coupled with the insulation properties, it could decrease the operational carbon emissions by conserving energy. This would lead to a pathway toward carbon neutrality.

(5) **Value-added products:** Despite the promising properties of 3D-printed wheat straw insulation structures, penetrating the market faces challenges. Conventional insulation products are not only low-cost to customers but also well-established in terms of performance. Establishing market and customer trust in eco-friendly alternatives requires not only superior properties but also strategic positioning. To this end, leveraging the unique capabilities of AM to produce complex and customizable structures, such as casts and footwear, offers a competitive edge. The emphasis should be on developing biomass materials that not only match but surpass the performance of traditional options, establishing themselves as compelling and sustainable alternatives within the evolving socio-technological market landscape.

(6) Resolution**:** During the DIW printing process, the size of the nozzle diameter determines the printing resolution. For example, higher resolution can be achieved by using a smaller diameter nozzle. However, due to the relatively large particles of wheat straw fiber powder, currently only a 1.54mm nozzle can be used. The mechanism of nozzle clogging during direct ink writing by Brendan[54] et al. Differences in fiber length, volume fraction, and nozzle geometry make clogging mechanisms diverse. Pores in the ink affect fiber orientation and thus plugging behavior. This means that the ink composition can be adjusted to prevent certain clogging mechanisms. By solving the clogging problem, the size of the nozzle can be smaller, improving the resolution of DIW printing.

4. CONCLUSION

This study marks a pioneering effort in the additive manufacturing of wheat straw fiber for thermal insulation. The direct ink writing process is a cost-effective manufacturing technology that provides a sustainable manufacturing solution. We evaluated the rheological properties of the prepared ink, mechanical properties of the freeze-dried and ambient pressure dried samples, and the thermal insulation performance of the 3D-printed straw fiber. The insights gained in this work provide a deeper understanding of the correlation between the material composition and the additive manufacturing process for sustainable insulation materials. This work lays the foundation for future optimizations in the manufacturing processes of straw fiber insulation materials. We also expect that this pilot work will serve as a blueprint for the broader adoption of diverse biomass-based sustainable materials, such as stalks, rice straws, leaves, or husks. Moving forward, the integration of sustainable material development and manufacturing technology will be crucial for addressing contemporary environmental challenges, such as carbon capture and sequestration, sustainability, and multi-functionalities.

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REFERENCES

[1] M. S. Al-Homoud, "Performance characteristics and practical applications of common building thermal insulation materials," *Building and environment,* vol. 40, no. 3, pp. 353-366, 2005.

[2] B. Abu-Jdayil, A.-H. Mourad, W. Hittini, M. Hassan, and S. Hameedi, "Traditional, state-of-the-art and renewable thermal building insulation materials: An overview," *Construction and Building Materials,* vol. 214, pp. 709-735, 2019.

[3] Y. Zhang, G. Zhou, K. Lin, Q. Zhang, and H. Di, "Application of latent heat thermal energy storage in buildings: State-of-the-art and outlook," *Building and environment,* vol. 42, no. 6, pp. 2197-2209, 2007.

[4] G. V. Research. "Global Building Thermal Insulation Market Size Report." https://www.grandviewresearch.com/industry-analysis/building-thermal-insulation-market (accessed.

[5] M. a. Markets. "Thermal Insulation Material Market by Material Type." https://www.marketsandmarkets.com/Market-Reports/thermal-insulation-material-market-189207197.html (accessed.

[6] S. Schiavoni, F. Bianchi, and F. Asdrubali, "Insulation materials for the building sector: A review and comparative analysis," *Renewable and Sustainable Energy Reviews,* vol. 62, pp. 988-1011, 2016.

[7] N. Ijjada and R. R. Nayaka, "Review on properties of some thermal insulating materials providing more comfort in the building," *Materials Today: Proceedings,* vol. 58, pp. 1354-1359, 2022.

[8] N. Llantoy, M. Chàfer, and L. F. Cabeza, "A comparative life cycle assessment (LCA) of different insulation materials for buildings in the continental Mediterranean climate," *Energy and Buildings,* vol. 225, p. 110323, 2020.

[9] F. T. Wallenberger and P. A. Bingham, "Fiberglass and glass technology," *Energy-Friendly Compositions And Applications,* 2010.

[10] L. Chen *et al.*, "Strategies to achieve a carbon neutral society: a review," *Environmental Chemistry Letters,* vol. 20, no. 4, pp. 2277-2310, 2022.

[11] C. G. Schirmeister and R. Mülhaupt, "Closing the carbon loop in the circular plastics economy," *Macromolecular Rapid Communications,* vol. 43, no. 13, p. 2200247, 2022.

[12] L. Lammert, "Circular economy in architecture: sustainable principles for future design," L. Lammert, 2018.

[13] P. Mindermann, M. Gil Pérez, J. Knippers, and G. T. Gresser, "Investigation of the fabrication suitability, structural performance, and sustainability of natural fibers in coreless filament winding," *Materials,* vol. 15, no. 9, p. 3260, 2022.

[14] S.-Q. Tian, R.-Y. Zhao, and Z.-C. Chen, "Review of the pretreatment and bioconversion of lignocellulosic biomass from wheat straw materials," *Renewable and Sustainable Energy Reviews,* vol. 91, pp. 483-489, 2018/08/01/ 2018, doi: https://doi.org/10.1016/j.rser.2018.03.113.

[15] V. Smil, "Crop Residues: Agriculture's Largest Harvest: Crop residues incorporate more than half of the world's agricultural phytomass," *Bioscience,* vol. 49, no. 4, pp. 299-308, 1999.

[16] C. Rojas, M. Cea, A. Iriarte, G. Valdés, R. Navia, and J. P. Cárdenas-R, "Thermal insulation materials based on agricultural residual wheat straw and corn husk biomass, for application in sustainable buildings," *Sustainable Materials and Technologies,* vol. 20, p. e00102, 2019.

[17] X. Yin, M. Lawrence, D. Maskell, and M. Ansell, "Comparative micro-structure and sorption isotherms of rice straw and wheat straw," *Energy and Buildings,* vol. 173, pp. 11-18, 2018.

[18] C. Koh, F. Gauvin, K. Schollbach, and H. Brouwers, "Upcycling wheat and barley straws into sustainable thermal insulation: Assessment and treatment for durability," *Resources, Conservation and Recycling,* vol. 198, p. 107161, 2023.

[19] Q. Cen, S. Chen, D. Yang, D. Zheng, and X. Qiu, "Full Bio-Based Aerogel Incorporating Lignin for Excellent Flame Retardancy, Mechanical Resistance, and Thermal Insulation," *ACS Sustainable Chemistry & Engineering,* vol. 11, no. 11, pp. 4473-4484, 2023.

[20] C. Wang *et al.*, "Cellulose as an adhesion agent for the synthesis of lignin aerogel with strong mechanical performance, Sound-absorption and thermal Insulation," *Scientific reports,* vol. 6, no. 1, p. 32383, 2016.

[21] H. Setyawan, M. a. Fauziyah, H. S. S. Tomo, W. Widiyastuti, and T. Nurtono, "Fabrication of Hydrophobic Cellulose Aerogels from Renewable Biomass Coir Fibers for Oil Spillage Clean-Up," *Journal of Polymers and the Environment,* vol. 30, no. 12, pp. 5228-5238, 2022.

[22] F. Zhang, C. Wang, C. Mu, and W. Lin, "A novel hydrophobic all-biomass aerogel reinforced by dialdehyde carboxymethyl cellulose for oil/organic solvent-water separation," *Polymer,* vol. 238, p. 124402, 2022.

[23] M. Lieder and A. Rashid, "Towards circular economy implementation: a comprehensive review in context of manufacturing industry," *Journal of cleaner production,* vol. 115, pp. 36-51, 2016.

[24] D. L. M. Nascimento *et al.*, "Exploring Industry 4.0 technologies to enable circular economy practices in a manufacturing context: A business model proposal,"

Journal of Manufacturing Technology Management, vol. 30, no. 3, pp. 607-627, 2019.
[25] J.-P. Schöggl, L. Stumpf, and R. J. Baumgartner, "The narrative of sustainability and circular economy-A longitudinal review of two decades of research," *Resources, Conservation and Recycling,* vol. 163, p. 105073, 2020.
[26] H. A. Colorado, E. I. G. Velásquez, and S. N. Monteiro, "Sustainability of additive manufacturing: the circular economy of materials and environmental perspectives," *Journal of Materials Research and Technology,* vol. 9, no. 4, pp. 8221-8234, 2020.
[27] A. B. L. de Sousa Jabbour *et al.*, "Circular economy business models and operations management," *Journal of cleaner production,* vol. 235, pp. 1525-1539, 2019.
[28] T. Brydges, "Closing the loop on take, make, waste: Investigating circular economy practices in the Swedish fashion industry," *Journal of Cleaner Production,* vol. 293, p. 126245, 2021.
[29] F. Blomsma *et al.*, "Developing a circular strategies framework for manufacturing companies to support circular economy-oriented innovation," *Journal of cleaner production,* vol. 241, p. 118271, 2019.
[30] A. Ben-Ner and E. Siemsen, "Decentralization and localization of production: The organizational and economic consequences of additive manufacturing (3D printing)," *California Management Review,* vol. 59, no. 2, pp. 5-23, 2017.
[31] L. Poudel, S. Elagandula, W. Zhou, and Z. Sha, "Decentralized and centralized planning for multi-robot additive manufacturing," *Journal of Mechanical Design,* vol. 145, no. 1, p. 012003, 2023.
[32] X. Bi and R. Huang, "3D printing of natural fiber and composites: A state-of-the-art review," *Materials & Design,* p. 111065, 2022.
[33] K. Oksman, M. Skrifvars, and J.-F. Selin, "Natural fibres as reinforcement in polylactic acid (PLA) composites," *Composites science and technology,* vol. 63, no. 9, pp. 1317-1324, 2003.
[34] K. Markstedt, A. Mantas, I. Tournier, H. Martínez Ávila, D. Hägg, and P. Gatenholm, "3D Bioprinting Human Chondrocytes with Nanocellulose–Alginate Bioink for Cartilage Tissue Engineering Applications," *Biomacromolecules,* vol. 16, no. 5, pp. 1489-1496, 2015/01/01/ 2015, doi: https://doi.org/10.1021/acs.biomac.5b00188.
[35] Z. Guo, R. Yang, T. Wang, L. An, S. Ren, and C. Zhou, "Cost-effective additive manufacturing of ambient pressure-dried silica aerogel," *Journal of Manufacturing Science and Engineering,* vol. 143, no. 1, p. 011011, 2021.
[36] Z. Guo, L. An, S. Lakshmanan, J. N. Armstrong, S. Ren, and C. Zhou, "Additive manufacturing of porous ceramics with foaming agent," *Journal of Manufacturing Science and Engineering,* vol. 144, no. 2, p. 021010, 2022.
[37] Z. Guo and C. Zhou, "Recent advances in ink-based additive manufacturing for porous structures," *Additive Manufacturing,* vol. 48, p. 102405, 2021.
[38] J. A. Lewis, "Direct ink writing of 3D functional materials," *Advanced Functional Materials,* vol. 16, no. 17, pp. 2193-2204, 2006.
[39] X. Song *et al.*, "Biomimetic 3D printing of hierarchical and interconnected porous hydroxyapatite structures with high mechanical strength for bone cell culture," *Advanced Engineering Materials,* vol. 21, no. 1, p. 1800678, 2019.
[40] M. Di Luigi, Z. Guo, L. An, J. N. Armstrong, C. Zhou, and S. Ren, "Manufacturing silica aerogel and cryogel through ambient pressure and freeze drying," *RSC advances,* vol. 12, no. 33, pp. 21213-21222, 2022.
[41] Q. Zhang, F. Zhang, S. P. Medarametla, H. Li, C. Zhou, and D. Lin, "3D printing of graphene aerogels," *Small,* vol. 12, no. 13, pp. 1702-1708, 2016.
[42] Z. Guo, F. Fei, X. Song, and C. Zhou, "Analytical Study and Experimental Verification of Shear-Thinning Ink Flow in Direct Ink Writing Process," *Journal of manufacturing science and engineering,* vol. 145, no. 7, p. 071001, 2023.
[43] A. Chami Khazraji and S. Robert, "Self-assembly and intermolecular forces when cellulose and water interact using molecular modeling," *Journal of Nanomaterials,* vol. 2013, pp. 1-12, 2013.
[44] Y. Li, M. Lin, and J. W. Davenport, "Ab initio studies of cellulose I: crystal structure, intermolecular forces, and interactions with water," *The Journal of Physical Chemistry C,* vol. 115, no. 23, pp. 11533-11539, 2011.
[45] R. J. Moon, A. Martini, J. Nairn, J. Simonsen, and J. Youngblood, "Cellulose nanomaterials review: structure, properties and nanocomposites," *Chemical Society Reviews,* vol. 40, no. 7, pp. 3941-3994, 2011.
[46] B. Medronho and B. Lindman, "Brief overview on cellulose dissolution/regeneration interactions and mechanisms," *Advances in colloid and interface science,* vol. 222, pp. 502-508, 2015.
[47] A. R. Abdullaev and I. N. Q. Zulxumor, "A Review On: Analysis Of The Properties Of Thermal Insulation Materials," *The American Journal of Interdisciplinary Innovations and Research,* vol. 3, no. 05, pp. 27-38, 2021.
[48] G. Wei, Y. Liu, X. Zhang, F. Yu, and X. Du, "Thermal conductivities study on silica aerogel and its composite insulation materials," *International Journal of Heat and Mass Transfer,* vol. 54, no. 11-12, pp. 2355-2366, 2011.
[49] F. Akhter, S. A. Soomro, and V. J. Inglezakis, "Silica aerogels; a review of synthesis, applications and fabrication of hybrid composites," *Journal of Porous Materials,* vol. 28, no. 5, pp. 1387-1400, 2021.
[50] T. Xie and Y.-L. He, "Heat transfer characteristics of silica aerogel composite materials: Structure reconstruction and numerical modeling," *International Journal of Heat and Mass Transfer,* vol. 95, pp. 621-635, 2016.
[51] J.-J. Zhao, Y.-Y. Duan, X.-D. Wang, and B.-X. Wang, "Radiative properties and heat transfer characteristics of

fiber-loaded silica aerogel composites for thermal insulation," *International Journal of Heat and Mass Transfer,* vol. 55, no. 19-20, pp. 5196-5204, 2012.
[52] M. Bogers, R. Hadar, and A. Bilberg, "Additive manufacturing for consumer-centric business models: Implications for supply chains in consumer goods manufacturing," *Technological forecasting and social change,* vol. 102, pp. 225-239, 2016.
[53] R. Martukanitz *et al.*, "Toward an integrated computational system for describing the additive manufacturing process for metallic materials," *Additive Manufacturing,* vol. 1, pp. 52-63, 2014.
[54] B. P. Croom *et al.*, "Mechanics of nozzle clogging during direct ink writing of fiber-reinforced composites," *Additive Manufacturing,* vol. 37, p. 101701, 2021/01/01/ 2021, doi: https://doi.org/10.1016/j.addma.2020.101701.