The Applications of Nanocellulose in 3D Printing

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Abstract:

3D printing was originally intended to realize mass production of various merchandise more conveniently and precisely, and it has achieved partly up to days. However, nowadays 3D -printed products still have some flaws, like inadequate shape stability and low printing precision. Meanwhile, nanocellulose, as a renewable natural material, is reckoned one of the most promising selections. This paper mainly highlights the utilizations and functions of nanocellulose in 3D printing. First, this paper offers a brief introduction concerning its structure, properties, and synthesis. Then, several practical applications in different domains are listed and anatomized to exemplify the necessity and superiority of the integration between nanocellulose and 3D printing. Moreover, this paper points out and generalizes some existing challenges and directions about 3D printing development and nanocellulose applications. Finite range of materials for printing still severely restricts the quality and performances of end-product. Lack of detailed standards to regulate and unify sources, dimensions, extraction and processing methods contributes to the miscellaneous specifications of nanocellulose. Consequently, consistent and profound refelction on cutting-edge technology and materials is essential for mankind and can also inspire us to utilize them in more rational, efficient and eco-friendly ways. The future of 3D printing and nanocellulose is still promising and anticipated to cover more industry by the integration with traditional manufacture.

Keywords: 3D printing; nano-cellulose; polymer; modified composite.

1. Introduction

3D printing, as a form of additive manufacturing (AM), refers to printing articles following digital

models through depositing numerous layers successively, achieving automatic and intelligent fabrication with higher efficiency and less waste. In 1986, Chuck Hull pioneered stereo lithography appearance (SLA),

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which marks the birth of 3D printing. As the technology progresses continuously, according to the different demands and materials, there are varieties of printing methods for selection, in which fairly mature ones include: SLA, fused deposition modeling (FDM) and selective laser sintering (SLS) [1]. Each of them has its particular advantages, applications and limitations. The concept of 3D printing makes structure designing more free and endows products with more complicated structures [2]. Products produced in this way have their unique advantages and become more in line with people's practical demands and expectations. Due to the features of 3D printing and further research and breakthroughs concerning it, 3D printing has been developed maturely enough to be extended into more domains, encompassing food [3,4], construction [5,6], aerospace [7], automotive [7,8], electronics [9,10] and medicine [11-13].

However, 3D-printed products are still confronted with drawbacks and challenges, especially compared with their counterparts fabricated by the traditional approaches. For example, the former tends to demonstrate worse strength and compatibility, anisotropy, low precision in printing and shaping, poor surface quality and relatively obvious printing marks [14,15]. In order to solve these problems, one feasible direction is to adjust printing parameters, improve or innovate existing techniques and design new 3D printers or introduce other technologies into 3D printing as supplementary means, in brief, making breakthroughs at the technical level. Multi-material printing and printers, applying unique material switching methods and structures, provide access to mix materials with different properties and textures simultaneously for one specific product [14,16]. Multi-axis 3D printing, perceived as an extension of traditional 3D printing, enables the print head to rotate more freely and flexibly, and thereby is helpful to fabricate complex microstructures and enhance the isotropy of products [17,18]. Multi-axis robot arms are introduced as assistance to achieve seamless multi-material printing [19]. Field-based volumetric 3D printing, instead of printing articles through the conventional layer-by-layer approaches, provides a brand new and distinct paradigm that permits revolutionary layerless printing through chemical reactions utilizing the responses of materials to assisted physical fields (such as optical and acoustic fields) [20]. Artificial intelligence (AI) and machine learning (ML) has collaborated with 3D printing for quality control, process optimization, design optimization, microstructure analysis and material formulation through intelligently and automatically analyzing, selecting and adjusting the optimal printing parameters and procedures [21]. Inspired by the principle of subtractive manufacturing, ultra-precision machining (UPM), as a post-processing procedure, is integrated with 3D printing, which enables to enhancement of the surface quality and shape accuracy of 3D-printed products [15]. The aforementioned methods are indeed conducive to ameliorating the quality and performances of the 3D-printed products, but they might make the whole printing system more sophisticated and cumbersome, elongate the whole printing process, reduce the whole efficiency, or even induce more technological problems.

Meanwhile, another probable solution is to innovate and modify the materials used in 3D printing, seemingly attempting to remedy the defects of properties and performances directly and, in other words, seeking possibilities from the perspective of materials. The kinds of materials suitable for 3D printing are diverse and still increasing, including polymers, ceramics, metals and their alloys, resins, other biocompatible materials (bioink) or advanced materials (smart materials like shape memory alloys and nanomaterials), and combinations of their hybrid, composite or functionally graded materials [7,14,22,23]. To our knowledge, polylactic acid (PLA) and acrylonitrile-butadiene-styrene (ABS) are the most common polymeric materials used in 3D printing. The former, despite its brittle and weak mechanical properties, is widely used through FDM because of its low melting point and biocompatibility, while the latter possesses excellent strength, physical and mechanical properties, better durability and heat, and impact resistance [23]. In sum, due to the requirements and necessity to improve the printing performances, mechanical properties, biocompatibility and sustainability of currently common 3D printing materials, some suitable and cost-effective additives or methods for reinforcements or modifications are in demand. Therefore, cellulose and its derivatives become probably viable substances.

Herein, nanocellulose is highlighted, and its properties and synthesis are introduced. Additionally, the theory of why it is beneficial to ameliorating the quality of 3D-printed articles is elucidated and several practical applications in different domains are summarized and listed. Finally, a discussion concerning the challenges and deficiencies, prospective and expectations of 3D printing and nanocellulose is given.

2. Structure, Properties and Synthesis of Nanocellulose

Nanocellulose, derived from cellulose, a sustainable and renewable biopolymer widely existing in nature, can be a potential solution to alleviate environmental concerns through replacing traditional organics and acting as a functional additive in polymer synthesis. With excellent physicochemistry properties, mechanical properties, lightweight-ness, abilities for chemical modification and

environmental friendliness, it has been used in several domains and industries, including food packaging, electronics, materials reinforcements, pharmaceuticals and the medical field [24].

Nanocellulose can be classified into three main kinds: cellulose nanofibers (CNF), cellulose nanocrystals (CNC) and bacterial nanocellulose (BNC). Among them, CNF and CNC are mainly obtained from various plants through a top-down process while BNC is mainly attained from living organisms [25].

CNF, whose length ranges from several hundred nanometers to several micrometers, is comprised of crystalline and amorphous regions. The synthesis of CNF mainly utilizes mechanical treatments which can produce high shear forces to decrease its dimension effectively. The methods include high-pressure homogenization, microfluidization, ball milling and ultrasonication. However, because of the hydrogen bonds existing between CNF and resultant aggregations, chemical pretreatment, like enzymatic hydrolysis and TEMPO-mediated oxidation, is proved to be necessary and useful. Additionally, these chemical pretreatments are conducive to reducing energy consumption during mechanical processing [26].

CNC, whose diameter and length range from 5 to 70 nanometers and from 100 to 250 nanometers, respectively, possesses a short rod-like or whisker-like shape, a high degree of crystallinity and excellent mechanical strength. Currently, the prevalent synthesis of CNC is acid hydrolysis. In acidic environments, the unstable amorphous regions in CNC are likely to be hydrolyzed under the attack of acid while only the crystalline regions with resistance to acid could remain. Then highly crystalline CNC could be separated through mechanical treatments. Moreover, sulfuric acid is the most commonly used because besides attacking amorphous regions, it can from sulfonic acid groups or sulfate half-ester groups with the hydroxyl groups on the surface of CNC, which endows the surface with electronegativity and enhances the dispersion stability of CNC in aqueous phase due to electrostatic repulsion [26].

BNC, whose diameter varies from 20 to 100 nanometers, is generated from microorganisms like Gluconacetobacter through a bottom-up way and therefore any pretreatments are unnecessary. With higher purity, better crystallinity, excellent mechanical properties and biocompatibility, BNC has been extensively used in the food industry, and biomedical and tissue engineering [25].

3. Practical Applications of Nanocellulose Concerning 3D Printing

In order to improve the comprehensive performances of

3D printing ink, specific eco-friendly and functional substances are in demand. Therefore, possessing large specific surface area, outstanding mechanical performances, biocompatibility and biodegradability, nanocellulose is one of the most suitable and promising materials used as an ingredient and functional modifier for 3D printing ink [27]. Herein, the effect of nanocellulose on 3D printing ink and the underlying mechanism will be explained.

3.1 Food Industry

Nanocellulose, due to its excellent mechanical performances, biodegradability and biocompatibility, is widely utilized as a reinforcement or barrier agent in food packaging or coatings [28]. In addition, it can be used as a modifier in some hydrogels to improve their rheological properties for 3D printing of food [29].

Zhou et al [30] inventively designed core-shell fibers and fabricated a functional label through coaxial 3D printing. As to the structure of their fibers, in the shell, CNF, sodium alginate (SA) and κ-carrageenan (KC) collectively comprised the matrix, and blueberry anthocyanins (BA) were used to monitor degree of freshness though color changes. In the core was the 1-methylcyclopropene (1-MCP) contained by chitosan, intended to be released under control to keep fruits or vegetables fresh. Among them, CNF significantly improved the rheological properties and shear thinning behavior of the ink by virtue of the higher shear rate and lowering viscosity, which enhanced the printability and fidelity of the ink. Moreover, such structure permitted the label with double functions including monitoring and refreshing. The sensitivity and accuracy of monitoring, and the feasibility of refreshing litchis for up to 6 days were proved through experiments. This work exemplifies the modification of nanocellulose on ink and the possibility of intelligent food packaging made by 3D printing.

Dermol et al [31] designed three different hydrogels with varying ratios of BNC to cationic starch (70/30, 60/40 and 50/50) and successfully fabricated packaging foils through their modified 3D printer following FDM method. The rheological performances of these hydrogels were improved due to BNC and the 70/30 sample displayed the best. In addition, a series of properties of these foils were measured. Foils with more BNC showed worse transparency and mechanical properties, while the foils printed by 50/50 hydrogel displayed comprehensive performances, including homogeneity, better transparency and higher rigidity. This work provides a viable solution to produce more sustainable and eco-friendly packaging foils through 3D printing.

Zhang et al [32] designed an innovative food-ink to produce some food that is easy to swallow and is suitable for

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dysphagia diet. Through experiments, two formulations were finally selected for 3D printing: 2wt% gelatin B (GB), 8wt% xanthan gum (XG), 1wt% CNC, and 8wt% whey protein isolate (WPI), 8wt% XG, 1.5wt% CNC. Adding CNC notably ameliorated the rheological properties, thermal stability and shape fidelity by forming hydrogen bonds and 3D network, allowing the printed food to maintain their original shape and be reserved at 4°C for at least 96h or even at 50°C. Moreover, according to the result of International Dysphagia Diet Standardization Initiative (IDDSI) tests, the printed sample conformed to the "level 5-minced and moist" dysphagia diet. This work indicates the possibilities and potential methods to proffer suitable, customized and edible food for those patients suffering dysphagia, and this sort of design thought could be extended to different patients or occasions.

3.2 Electronic Industry

Patel et al [33] integrated nanocellulose and β-glucan (BG) with chitosan methacrylamide (CSM) to synthesize one new kind of multifunctional hydrogel, followed by discussing its performances, possibilities of being utilized as sensors and moist electric generators. First, compared with the pure polymer hydrogel of CSM, this composite hydrogel was printable, more viscoelastic, electro-conductive, biocompatible, bio-adhesive and possessed distinct abilities like shape memory and self-healing, partly due to the reinforcement and modification of nanocellulose. In addition, the characteristic length of this composite hydrogel (187.46 nm) was shorter than that of pure polymer hydrogel (195.03 nm) because nanocellulose contributed to the self-assembly process of the hydrogel. As to potential applications, it displayed real-time motion-sensing or strain-sensing ability, and moisture-mediated electricity phenomenon. The former was assessed on different human body parts and the experimental results (waveforms) were nearly identical to the measurements, which manifested its accuracy. The latter was observed through thermo-responsive features which are embodied in the systematic variations of current caused by the rise and fall of temperature. This work inspires us to prepare hydrogels with functional and biocompatible materials from nature for 3D printing bioink or portable and wearable electronic devices for customized medical care.

Zhou et al [34] utilized a series of different CNF as modifiers to improve the printability, rheological performance of the MXene-based ink. This modified hybrid ink endowed products with better shape fidelity and printing accuracy. Particularly, CNF also prevented MXene nanosheets from restacking. That was why these pectinate electrodes fabricated through 3D printing and the freeze drying process, possessed multi-layered porous structures

which could enhance their ion migration efficiency and capacitive performance. In addition, based on this discovery, a solid-state supercapacitor with many excellent electrochemical performances was also printed. It performed a volumetric capacitance of 25.4F cm⁻³ at 1 mA cm⁻², displayed an energy density of 101 µWh cm⁻² at a power density of 0.299 mW cm⁻² and maintained a capacitance retention rate of 85% over 5000 cycles. This work demonstrates the feasibility of applying nanocellulose as rheological modifiers for MXene-based ink and the prospect of 3D-printed energy storage devices with distinct internal structures that are beneficial to electronic performances. Du et al [35] utilized CNF, lignin derived carbonized nanotube (LCNT) as ingredient and facile extrusion 3D printing (DIW) to fabricate conductive composite aerogel and its further application like pressure sensors. This kind of composite aerogel showed excellent mechanical performance. Among them, the sample LCNT/CNF-15(CNF/ LCNT ratio in weight was 85:15) maintained a stress retention of 92.5% with a constant 30% compression strain exerted for 1000 cycles. Moreover, after freeze drying process, this sample possessed a homogeneous and porous internal structure, and therefore was applied to produce a piezoresistive type pressure sensor. Through experiments, with testing pressure ranging from 0.2 to 9.8kPa, the relatively stable signal could be received and the response time was between 100 and 200 ms, which demonstrated its accuracy and sensitivity. This work provides us a potential design thought to fabricate wearable pressure sensors through composite aerogel mainly made of sustainable nature materials like nanocellulose and lignin.

4. Challenges and Perspectives

First, for 3D printing, on one hand, its techniques and methods have been diverse and each of them has specific application fields. However, their corresponding materials and printers are different from each other, resulting in fairly isolated and complicated selections. Furthermore, the development of this technology and the design of its apparatuses tend to be more sophisticated, making them more difficult to popularize and universalized for individuals or families.

On the other hand, compared to the conventional manufacturing industry, the materials for choosing are still limited and the processing speed of 3D printing is uncompetitive. Materials in use cannot be fully employed and several potential materials with great properties could not be utilized for 3D printing, both because they are not suitable enough for 3D printing and confined by some factors, like form and temperature. There is an ideal thinking that 3D printing should be perceived as a supplementary and

auxiliary means to collaborate with conventional manufacture, instead of superseding each other.

In sum, future research on 3D printing should take into consideration the extension and spreading towards society and personal usage, instead of only centering in advanced domains for enterprises or institutes. Meanwhile, we should also pay attention to the impact on the environment and society. To achieve this future vision, 3D printing technology is supposed to be gradually simplified, and highly integrated, and materials should be more affordable, sustainable, biodegradable, and recyclable.

Concerning nanocellulose, attributed to its microstructure, and prominent mechanical and chemical performances, it is indubitable that it is indeed a kind of sustainable ingredient, feasible modifier and reinforcement for 3D printing ink, which can enhance the quality and functions of printed articles, and endow them with unexpected properties or frameworks. Based on this, we have successfully discovered and exploited its potential to utilize it in several domains as a versatile, but perhaps there is still something latent for us to unveil.

Notwithstanding, we are still puzzled by some stingy problems. First, the dimensions, properties, and costs of different nanocellulose can be variable, since it can be derived from a plethora of natural sources. In addition, different extraction techniques and pretreatment methods also influence those aforementioned aspects. Therefore, stable supply and sources of raw cellulose, relatively cheap and time-saving methods for processing, or even some more standardized and commercialized nanocellulose should be propounded and provided as soon as possible.

5. Conclusions

This paper mainly introduces and summarizes the overview of nanocellulose, and its effect in 3D printing or applications with 3D printing.

Nanocellulose has three types and they are different from each other in structure and synthesis, which endows them with specific properties. Especially, owing to the mechanical performances and biocompatibility, nanocellulose has been used as an additive, modifier, or reinforcement for 3D printing ink. The main purpose of adding nanocellulose is to increase the sheering rate and to significantly ameliorate the rheological performances of ink. As a result, the ink is easier to flow and extrude from the nozzle of the printer, and the comprehensive behavior of printed articles will be more satisfactory. Several applications in the food industry, electronic devices and other applications are listed and analyzed.

Finally, the current challenges and prospects of 3D print-

ing and nanocellulose materials are given respectively. 3D printing technology needs to develop consistently to break the limitations of materials, and its relatively mature parts are supposed to be more popularized and affordable. Nanocellulose should have a standard concerning its source, dimension, and properties and should be extended into further sectors.

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