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Conductive Polymer Composites Based Flexible Strain Sensors by 3D Printing: A Mini-Review

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With the development of wearable electronic devices, conductive polymer composites (CPCs) based flexible strain sensors are gaining tremendous popularity. In recent years, the applications of additive manufacturing (AM) technology (also known as 3D printing) in fabricating CPCs based flexible strain sensors have attracted the attention of researchers due to their advantages of mold-free structure, low cost, short time, and high accuracy. AM technology, based on material extrusion, photocuring, and laser sintering, produces complex and high-precision CPCs based wearable sensors through layer-by-layer stacking of printing material. Some high-performance CPCs based strain sensors are developed by employing different 3D printing technologies and printing materials. In this mini-review, we summarize and discuss the performance and applications of 3D printed CPCs based strain sensors in recent years. Finally, the current challenges and prospects of 3D printed strain sensors are also discussed to provide an insight into the future of strain sensors using 3D printing technology.

Keywords: 3D printing, nanocomposites, strain sensor, nanofillers, performance

INTRODUCTION

In recent years, CPCs have been widely used in industrial and academic fields because of their flexibility, controllable conductive properties, easy processing, and low-cost synthesis (Wu et al., 2016; Wang T. et al., 2018; Li et al., 2018). Nanofillers are introduced to the insulating polymer matrix to improve their functional characteristics (Wang et al., 2018b). Due to the superior properties of functionalized CPCs, they are used to make strain sensors which are playing their role in medical diagnosis, soft robotics, health monitoring, human-computer interaction, and many other applications (Wang et al., 2018c; Wang et al., 2019; Zhang et al., 2019). Several methods for fabricating the strain sensors have been developed, such as hot pressing, casting, and extrusion. However, the processing technologies of these methods are complicated and not applicable to complex structures. For example, customized molds are needed to fabricate CPCs based strain sensors by hot pressing method (Huang et al., 2021). The development and production of mold limit the economic benefits during their industrial-scale production.

Since the last decade, AM technology, also known as 3D printing Zhang J. et al. (2020), is used by researchers for the fabrication of CPCs based strain sensors (Zhang et al., 2016; Gnanasekaran et al., 2017; Dawoud et al., 2018). Traditional fabrication methods used for preparing the molds of complex 3D objects are highly inefficient and time consuming. Compared with the conventional fabrication methods, 3D printing technology has the advantages of high resolution, high accuracy, low cost, fast

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TABLE 1 | The principle and characteristics of 3D printing.

Process	Materials	Principle	Advantages	Disadvantages
FFF	Thermoplastics	Fusion-driven restacking	Fast printing low cost High strength	Pretreatment Weak interaction Rough surface
DIW	Thermoplastics Elastomers Hydrogels	Ink extrusion	Material diversity Low cost	Nozzle clogging Post-processing
DLP	Photosensitive resin	*In situ* photocuring	High resolution Intricate fabrication	Weak strength High cost
SLS	Thermoplastics	Laser sintering	Material diversity High efficiency High material utilization	Rough surface Powder pollution High cost

TABLE 2 | 3D-printed strain sensors.

Methods	Printed materials[a]	Printed parts	Maximum strain	Gauge factors (GF)	Ref
FFF	Modified CNT/TPU	Entire sensor	250%	117213	Xiang et al. (2019)
FFF	CNT/TPU	Entire sensor	100%	8.6–176	Christ et al. (2017)
FFF	CNT/GNP/TPU	Entire sensor	250%	136327.4	Xiang et al. (2020a)
FFF	Graphene/TPU	Entire sensor	200%	15.2–155.7	Gul et al. (2019)
FFF	Modified CNT/TPU	Entire sensor	30%	3–130.9	Xiang et al. (2020c)
FFF	AgNPs/CNT/TPU	Entire sensor	250%	43260	Xiang et al. (2020b)
DIW	PDMS	Substrate	30%	4.3	Abshirini et al. (2018)
DIW	EGO/PDMS	Entire sensor	40%	20.3	Shi et al. (2019)
DIW	Graphene/PDMS	Sensing layer	350%	18.5–88443	Ma et al. (2019)
DIW	Nanoclay/CB/TPU	Entire sensor	80%	0.39–4.7	Wei et al. (2019a)
DIW	Ag precursor	Sensing layer	50%	—	Song et al. (2017)
DIW	Ag-TPU	Entire sensor	>1600%	—	Sun et al. (2019)
DLP	CNT/EA	Entire sensor	60%	1.587–8.939	Xiao et al. (2020)
DLP	c-CNTs/ACMO	Entire sensor	100%	2	Guo et al. (2020)
DLP	ACMO/HUA	Substrate	150 kPa	0.005–0.111kPa^{-1}	Peng et al. (2020b)
SLS	Graphene/TPU	Entire sensor	15%	668.3	Mei et al. (2020)

[a] *ABS, acrylonitrile butadiene styrene; ACMO, N-acryloyl morpholine; Ag, silver; AgNPs, silver nanoparticles; CB, carbon black; CNT, carbon nanotube; CNTs, carboxyl carbon nanotubes; c-HUA, hydrolyzable crosslinking agent; EA, elastomer; EGO, electrochemically derived graphene oxide; GNP, graphene nanoplatelet; PDMS, poly (dimethylsiloxane); TPU, thermoplastic polyurethane.*

fabrication, and customized for mass production without the need for mold (Abshirini et al., 2019; Dul et al., 2020; Song et al., 2020). Furthermore, due to the micro-orientation in Fused Filament Fabrication (FFF) and Direct Ink Writing (DIW) 3D Printing, the nanofillers are uniformly dispersed in the polymer matrix, leading to the enhanced performance of obtained strain sensor (Guo et al., 2021). Therefore, 3D printing through layer-by-layer manufacturing has shown promising potential to produce flexible strain sensors with complex structures. Their application areas include wearable electronic devices, medical diagnostics, soft robots, and strain direction recognition (Wang et al., 2020; Dan et al., 2021; Gao et al., 2021). Although 3D printing has been widely used in the development of complex structures, it is still a novel method to fabricate strain sensors with complex structures and multiple functions.

At present, there are some reports on the fabrication of CPCs based strain sensors with high performance by 3D printing. Considering the wide application potential of 3D printing in CPCs based strain sensors, it is necessary to review this topic. In this mini-review, 3D printing technologies based on material extrusion, photocuring, and laser sintering, involving Fused Filament Fabrication (FFF), Direct Ink Writing (DIW), Digital Light Processing (DLP), and Selective Laser Sintering (SLS) are discussed. Moreover, high-performance CPCs based strain sensors fabricated by these 3D printing technologies are reviewed. Further, the printing principle, material selection, research progress, recent applications, and prospects of CPCs based 3D printed strain sensors are also discussed.

FABRICATION OF CPCS BASED FLEXIBLE STRAIN SENSORS BY 3D PRINTING

3D printed CPCs based flexible strain sensors involve four different fabrication approaches including FFF, DIW, DLP, and SLS. The summary of materials, printing principles,

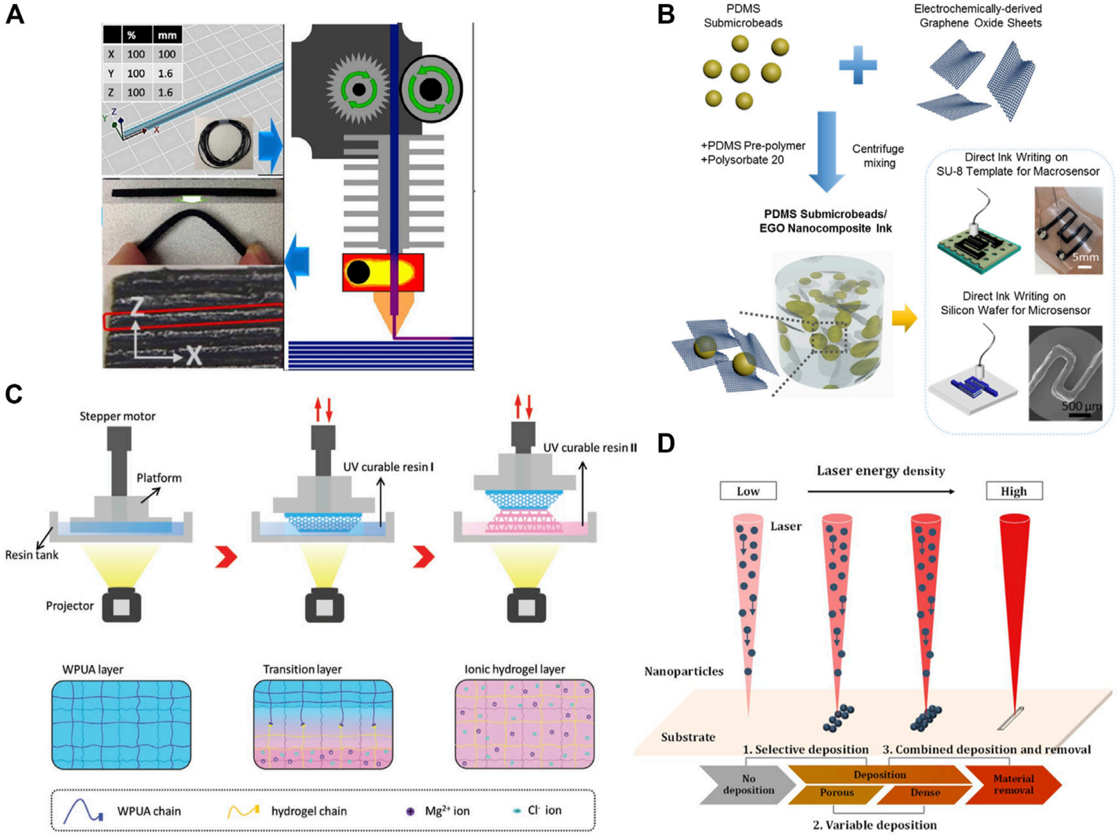

FIGURE 1 | The process for preparing strain sensor using **(A)** FFF (Christ et al., 2017), **(B)** DIW (Shi et al., 2019), **(C)** DLP (Yin et al., 2019), and **(D)** SLS (Song et al., 2020).

advantages, and disadvantages of these technologies is enlisted in **Table 1**; **Table 2** summarizes the representative researches of strain sensors fabricated by 3D printing in recent years.

Electrical properties of elastic polymer material improve with the introduction of conductive nanofillers. Furthermore, due to the increase of the interface density of nanofillers Arif et al. (2020) and the interaction between nanofillers and polymer matrix (Xiang et al., 2019), the interface between them is well combined, which facilitates a effective stress transfer under strain. Therefore, the mechanical properties of the composites, including strength, Young's modulus and toughness, are enhanced. (Pan et al., 2020). In addition, the sensing properties such as strain detection range, sensitivity, linearity, response time, resistance responsiveness, stability, and durability under different conditions are equally significant for CPCs based strain sensors. Material type, agglomeration and dispersion of nanofillers, macroscopic and microscopic structure of strain sensors, and different 3D printing parameters (Davoodi et al., 2020; Xu et al., 2020) determine the percolated network structure of the strain sensor, affecting the sensing properties of strain sensor. For example, when the percolation networks are in high density, numerous nanofillers contact with each other. The contact resistance is dominant and a robust percolation network is built. With the decrease of percolation networks density, more nanofillers are separated from each other, and contact resistance is gradually transformed into tunneling resistance. It makes the contact nodes between nanofillers easier to be disconnected, resulting in a higher sensitivity of composite. When the percolation networks are at low density, most of nanofillers could be in the state of tunneling effect. Here the tunneling resistance is dominant, leading to a fragile percolation network, which makes the sensitivity of composite increase sharply (Hu et al., 2012).

Flexible Strain Sensors Fabricated by FFF

Figure 1A illustrates the process for preparing strain sensor using FFF (Christ et al., 2017). The matrix material usually determines the stretchability of the fabricated strain sensor (Waheed et al., 2019; Alam et al., 2020; Arif et al., 2020), and TPU is undoubtedly a promising candidate for fabricating FFF based strain sensors because of its high strength, good elasticity and environmental resistance (Georgopoulou et al., 2020a). However, the lower dispersion of conductive nanofillers in these strain sensors leads to poor sensing performance.

The performance of 3D printed strain sensors could be enhanced through a synergistic effect between the nanofillers and increasing interaction between nanofillers and matrix to improve the dispersion of nanofiller in matrix (Georgopoulou and Clemens, 2020b). Xiang et al. (2020b) used FFF to prepare AGNP/CNT/TPU strain sensors with a synergistic effect, where AGNPs interact with CNTs. Micro-orientation (Guo et al., 2021) due to CNTs rearrangement during printing and synergistic effect between AGNPs and CNTs results in a wide strain detection range (0–250%), high sensitivity (GF = 43,260 at 250% strain), linearity ($R^2 = 0.97$ within 50% strain) and fast response (~57 ms). Synergies between other nanofillers, such as CNTs and GNPs have also been reported to achieve high performance (Xiang et al., 2020a). 1-pyrene carboxylic acid (PCA) was used to modify the TPU/CNT composite, and the dispersion of CNTs in composites was improved by applying the non-covalent interactions between PCA, TPU, and CNTs (Xiang et al., 2019). A 3D-printed piezoresistive strain sensor with a complex structure was also fabricated by non-covalent modification (Xiang et al., 2020c).

Compared with the conventional methods, FFF has the advantages of rapid synthesis (Christ et al., 2017), improved material utilization (Gnanasekaran et al., 2017), simple post-processing, no harmful by-products to the environment (Xiang et al., 2020b), and custom-made mass production without the need of molds (Dul et al., 2020). However, FFF also has some disadvantages: before printing, the prepared composite needs to undergo preprocessing, including granulation of composite and filament extrusion (Xiang et al., 2019). In addition, the printed components have a rough surface and limited physical interaction between adjacent filaments that reduce the structural strength along the deposition direction (Waheed et al., 2019).

Flexible Strain Sensors Fabricated by DIW

Ink extrusion is an effective method to print high-performance strain sensors. Inks have unique rheological properties, including viscoelasticity, shear-thinning, and yield stress, which contribute to the implementation of the DIW process. Usually, DIW is used to print the core components of the sensor or the template to prepare the sensor Wang Z. et al. (2018) and to meet the versatility of test environments. The stretchability of CPCs based strain sensors varies dramatically with the types of polymer materials (Sun et al., 2019; Wei P. et al., 2019). Particularly, PDMS elastomers are widely used in DIW printed flexible strain sensors because of their high stretchability, physiological inertia, controllable viscosity, and room temperature curing ability (Abshirini et al., 2019). **Figure 1B** illustrates the process of preparing the EGO/PDMS strain sensor by DIW (Shi et al., 2019). However, strain sensors fabricated by DIW using PDMS elastomers have lower sensitivity.

The sensitivity of the strain sensors is improved in two ways. The first is to optimize different printing parameters such as printing times (Song et al., 2017), which affects the degree of conductive network constructed in strain sensors, thus affecting the sensitivity. The other is the construction of a different microstructure. The conductive network of strain sensors, with different microstructures, changes under strain and controls the sensitivity of sensors. For example, a self-compensating two-order strain sensor was fabricated by Ma et al. (2019) through DIW and coating that revealed high GF (from 18.5 to 88 443) in a strain range of 0–350%. These results demonstrated that graphene coated on the surface of the sensor provides an additional conductive path, which self-compensates the loss of conductivity under high strain. Moreover, due to the slip-off and disconnection (Zhang X. et al., 2020) of internal graphene sheets, reduced graphene layers improve the sensitivity under small strain.

In addition, the performance of the flexible strain sensor improves by introducing non-conductive fillers to the CPCs. The immiscible second phase fluid was introduced to Ag/PDMS and Ag/TPU inks to fabricate the strain sensor by DIW (Sun et al., 2019). Due to the capillary action and the connection between the Ag sheets and the immiscible second phase fluid, the strain sensor achieves ultra-stretchability (>1,600%). Moreover, the shear flow generated in DIW rearranges and redistributes the conductive nanofillers (Huang et al., 2018), which results in the better orientation and uniform dispersion of the conductive nanofillers in strain sensors.

Unlike FFF technology, which applies only to thermoplastics, DIW is not limited to specific materials and is suitable for a variety of materials such as metal-based materials (Kim et al., 2019), carbon-based materials (Abshirini et al., 2018), and conductive polymer composites (Shi et al., 2019). However, DIW printing technology also has certain limitations; for different inks with different components, the print pattern requires post-processing such as soaking, sintering, heating, and curing (Khosravani and Reinicke, 2020). In addition, the processing resolution of the DIW printed strain sensors is less than that of FFF.

Flexible Strain Sensors Fabricated by DLP

Photocurable 3D printing is an emerging 3D printing method for wearable electronic devices. In this technique, ultraviolet light illuminates the mixture of photoinitiators, monomers, and prepolymers, initiating layer-by-layer curing according to the software-designed structure.

Photosensitive resins typically include acrylics, polyesters, and polyurethane acrylate containing composite precursors. Amon these, acrylic-based composites demonstrate fragile mechanical strength (Liu et al., 2021). To solve this problem, researchers used reactive diluents of acrylic-based composites to improve the mechanical strength as well as the transparency of composites (Peng et al., 2020a). Some studies report significant improvement in the mechanical stability of DLP-based strain sensors through the interlayer chemical bond formation between two materials, and the preparation process is shown in **Figure 1C** (Yin et al., 2019). The sensor exhibits excellent structural stability and sensing performance (10,000 loading/unloading cycles) due to the chemical bonds between hydrogel and WPUA. On one hand, the mechanical properties of photosensitive resins could improve by adding nanofillers (Mu et al., 2017; Cortes et al., 2020). On the other hand, the addition of nanofillers may absorb ultraviolet light, leading to the decline of the mechanical properties of the photosensitive resin.

In many studies, the strain detection range of DLP-based strain sensors is low (less than 30%) Cortes et al. (2020) and difficult to meet the practical application requirements. To solve this problem, Xiao et al. (2020) fabricated a flexible strain sensor based on CNTs/epoxy aliphatic acrylate (EAA)/aliphatic urethane diacrylate (AUD) composites and achieved a maximum strain detection range of 60%. Guo et al. (2020) introduced carboxyl CNTs to ACMO resin, which fully mitigated the over-curing of ACMO resin, thus, achieved a 100% strain detection range. Peng et al. (2020b) prepared a strain sensor with high stretchability (~510%) by DLP printing sacrificial mold method.

These strain sensors have low sensitivity that does not respond to subtle strains. Peng et al. (2018) used DLP to customize the microstructure (pyramid, semisphere, and semicylinder) template and to prepare the strain sensor with high sensitivity (maximum GF of $-3.6\ \text{kPa}^{-1}$). At present, the research on strain sensors with a high strain detection range and high sensitivity is still scarce, and most of the designed sensors only achieve sensitivity or detection range. For DLP-based strain sensors, more research needs be done in the future to achieve high performance.

Compared with FFF and DIW based 3D printing, DLP overcomes the pre-processing and post-printing processing and has higher efficiency. Besides, the sample printed by DLP has a high resolution, indicating its higher precision (Liu et al., 2021). However, DLP is limited to the printed materials, only applicable to photosensitive resins, and has a high printing cost, thus limiting the development of DLP technology. In addition, to decrease the resin viscosity and the ultraviolet absorption of nanofillers is challenging for the fabrication of photosensitive composite resins.

Flexible Strain Sensors Fabricated by SLS

Many flexible SLS-based strain sensors have been studied in recent years (Maier et al., 2013). The performance of the strain sensor can be improved by optimizing the printing parameters of SLS and constructing a special conductive network structure. The printing parameters are adjusted to make the conductive network structure in strain sensor more completed, as shown as **Figure 1D**; (Song et al., 2020). Zhuang et al. (2020) prepared CNT/TPU sensors with a wide range (17–240 kPa) and high sensitivity (0.12–0.549 kPa^{-}1) through SLS and improved the pressure detection range (3–240 kPa) and sensitivity ($1.357\ \text{kPa}^{-1}$ for 3–70 kPa, $0.0328\ \text{kPa}^{-1}$ for 70–240 kPa) by adjusting the SLS laser power, layer thickness, and scan spacing. (Zhuang et al., 2021). A segregated structure was constructed by SLS to make the nanofillers selectively distributed on the matrix surface. (Wei S. et al., 2019; Ronca et al., 2019). The segregated structure enables the sensor to achieve a low percolation threshold (0.05 wt%) (Gan et al., 2019). In addition, the synergistic effect improves the sensing performance of SLS-based strain sensors (Rollo et al., 2020).

Comparing the 3D printing methods discussed above, SLS has the highest production efficiency, and the material utilization rate is about 100%. However, powder pollution in the printing process, high operating costs, high equipment cost, and rough product surface are few problems associated with SLS. In addition, the printed product is deformed due to the long storage time and internal stress.

APPLICATION

3D printed high-performance strain sensors are applied in many fields such as medical diagnosis and health monitoring (Khosravani and Reinicke, 2020; Liu et al., 2021). For monitoring human movements, 3D printed strain sensors have a wide range of applications (Le et al., 2017), such as finger bending, wrist bending, elbow bending, walking, and running (Christ et al., 2018). In health care systems, 3D printed strain sensors monitor patients' respiratory and pulse rates, speech recognition, gestures, and blinking (Kim et al., 2017). Another potential application of 3D printed sensors is load recognition, including load distribution of heavy objects and light objects (Zhang et al., 2019). Besides, the high sensitivity strain sensors are also used to detect subtle acoustic vibration signals (Song et al., 2017). When the sensors are stimulated by external forces or sound waves, vibrations increase the sensing resistance patterns, and convert mechanical energy into electrical signals. In addition, DLP-based 3D printing combined with the self-pinning carbon nanotubes and assisted by out-of-plane capillary forces, the prepared strain sensor detects out-of-plane forces (Liu et al., 2018). In general, the 3D printed strain sensors have a wide range of applications in production, life, and scientific research (Bekas et al., 2019).

DISCUSSION

Among different methods for preparing the CPCs based strain sensors, extrusion, photocurable, and laser sintering 3D printing are rapid, precise, customized, and economical, compared to traditional methods. But these methods have certain drawbacks. FFF printing requires pretreatment of the composite material and filament manufacturing. During the FFF printing nozzle blockage and roughness of the surfaces of the finished product are other challenges. Due to the different compositions of the printing ink in the DIW process, the nozzle gets blocked. Moreover, the resolution of DIW is relatively low. DLP has a high manufacturing cost and is limited to photosensitive resins only. In addition, powder pollution in the printing process, high operating costs, high equipment cost, and rough product surface are few problems associated with SLS. Given the above shortcomings, combined with the challenges associated with industrial production, it is necessary to explore new 3D printing methods with high precision, high efficiency, and high applicability.

The development of wearable electronics is rapid, resulting in more electronic product waste and non-decomposable materials, which increase the pressure on the environment. Therefore, the development of recyclable, biodegradable, and biocompatible materials as 3D printing materials is the trend of 3D printed CPCs based strain sensors.

At present, most of the 3D printed CPCs based strain sensors have a single function. With the increasing demand for flexible strain sensors and their wide applicability, the realization of their multi-functional use is also a trend for the future development of 3D printed CPCs based strain sensors.

CONCLUSION

As a preparation method for flexible strain sensors, 3D printing technology is economical, rapid, highly precise, and has been widely used in industrial and academic fields. In this mini-review, we summarize the printing principle, material selection, advantages and challenges of 3D printing. Based on the research progress, the methods, involving synergistic effect, non-covalent modification, printing parameters, the construction of microstructure, are summarized to improve the strain detection range and sensitivity of 3D-printed strain sensors. Furthermore, 3D-printed strain sensors are used in human motion detection, health care systems, direction recognition and other fields. The challenges and future prospects are put forward. There are still some challenges to be solved to achieve commercialization, the development of 3D printing technology will continue to support the preparation of flexible strain sensors, which will make 3D printing an indispensable technology in the field of wearable electronic devices.

AUTHOR CONTRIBUTION

LL contributed to the compilation of data and the writing of the Manuscript. DX and ZZ contributed to the review, revision and editing of the Manuscript. YW, YL, HL, and CZ contributed to the review, revision of the Manuscript. All authors read and agree to the final text.

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