Study on the forming and sensing properties of laser-sintered TPU/CNT composites for plantar pressure sensors

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Abstract

Conductive polymer composites (CPCs) combining with specific microstructures (micropores, microcracks, etc.) can exhibit unique resistance response changes, which can be widely regarded as an effective way to improve sensing performance. This study takes advantage of the characteristics of the formation of tiny pores between crystal grains during selective laser sintering (SLS) processing to introduce a microporous structure into the thermoplastic polyurethane (TPU)/carbon nanotube (CNT) sensing element to prepare a three-dimensional porous conductive structure. The effect of the SLS process on sensing sensitivity, accuracy, and density was studied, and its sensing and forming mechanism were discussed. By adjusting SLS process parameters to control the performance of porous structure sensor elements, a final TPU/CNT sensor element with a wide pressure detection range, high sensitivity, a fast response time, and good stability and durability was developed. Finally, the optimal performance of the developed flexible pressure sensor was successfully used to detect the pressure distribution of the human foot. This study provided a simple and effective research method to develop high-performance flexible pressure sensors.

Keywords Selective laser sintering · Pressure-sensing element · Thermoplastic polyurethane · Carbon nanotubes · Conductive polymer composites

1 Introduction

Recently, novel resistive-type conductive polymer composite (CPC)-based strain sensors have attracted attention based on their merits of light weight, flexibility, stretchability, and easy processing, thus showing great potential applications in the fields of software robots [1], electronic skins [2], smart wearable devices [3], human movement detection [2, 4, 5], etc. Owing to the flexibility and stretchability of the polymer matrix, CPCs also have great potentials for the detection of various external stimuli (tensile, compression, organic vapor, temperature, etc.) based on the reconstruction of conductive

networks, which induced significant resistance variation [6–8]. Generally, CPCs are fabricated through combining conductive fillers (Ag nanowire, carbon nanotube [9], graphene [10], etc.) and insulating polymers with proper processing technology (melt mixing, solution mixing, dip-coating, spray coating, etc.).

Carbon nanotubes (CNTs) are often used as conductive filler materials and are good materials for preparing CPC-based strain sensors because of their large specific surface area, aspect ratio, high specific strength and modulus, and hardness and toughness [11, 12]. Michelis et al. [13] built a highly reproducible and hysteresis-free flexible strain sensor using inkjet printing CNTs on ethylene tetrafluoroethylene sheets. Liu et al. [14] prepared a lightweight porous thermoplastic polyurethane (TPU)/CNT sensor using the thermally induced phase separation (TIPS) technology, which has ultra-high compressibility and fast sensing capacity over a wide strain range (up to 90%). Lee et al. [15] applied silicone rubber to polyurethane foam and further dip-coated it with multiwalled carbon nanotube (MWCNT) dispersed TPU ink to produce a sensor that has good repeatability, high durability, and fast response speed, and the sensor can measure pressures of less than 100 Pa and more than 200 kPa. However, the

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surfaces of these CPC-based strain sensors are generally planar, as they are formed by hot compression and casting in a specific mold, which means that these pressure sensors cannot completely fit objects with complex curved surfaces, resulting in measurement errors.

Additive manufacturing (AM) is currently the fastest-growing manufacturing method. It allows the manufacturing of products with complex geometry and high dimensional accuracy, as well as high strength [16]. Following the increasing interest in AM, some AM technologies such as stereolithography, fused filament fabrication, and fused deposition modeling technologies are gradually popularized. Nevertheless, this approach has a limited range of applications due to the relatively low mechanical strength of applied polymer materials and several technological restrictions [17–20]. Recent development of AM methods, i.e., selective laser melting, electron beam melting, selective laser sintering (SLS), and laser engineering net shaping (LENS), offers new possibilities to conduct investigations on complex surfaces and lattice structures. Selective laser melting and LENS technologies use a high-power fiber laser with a wavelength of 1.064 μm as the energy source; these technologies are more suitable for the absorption and melting of metal alloy powders. Selective laser sintering uses a CO_2 laser with a wavelength of 10.64 μm , which is more suitable for sintering thermoplastic polymers, coated sand, and ceramic powder. Additionally, SLS is a wide range of processing materials and no support material is needed [21, 22], which make it more suitable for preparing flexible pressure-sensing elements with a complex curved surface structure. Among the commonly used polymer materials for laser sintering, TPU is an excellent electrical insulator that exhibits high elasticity, flexibility, and environmental stability. It is widely used in textile, medical, automotive, aerospace, aviation, and other fields, and it is an ideal base material for developing CPCs. Additionally, due to the unique forming process mechanism of laser sintering, it is possible to produce inherently porous objects. These pressure-sensing elements with a porous structure can greatly improve the sensitivity and gauge coefficient of the sensing element [23].

Laser sintering is a very complex thermophysical process, which depends not only on the accuracy of the SLS equipment and the thermophysical parameters of the material but also on the process parameters of the SLS [24, 25]. For commercial SLS equipment, only a few process parameters, such as laser power (P), layer thickness (h), scanning speed (v), and scan spacing (b), are relatively easy to control and adjust. The three parameters (P , v , and b) need to cooperate to achieve the energy ($E = P/(v \cdot b)$) required for powder sintering. Additionally, the sensitivity of the pressure sensor element studied in this paper is affected not only by the material but also by the porosity of the sensor element. The scan spacing affects the density in the horizontal direction (XY plane), and the layer thickness affects the density in building direction (Z).

Therefore, this study uses laser power, layer thickness, and scan spacing as input parameters for experiments. In the research of laser processing and molding optimization, many scholars adopted different experimental methods. Flores et al. [26] used the topology optimization method to optimize the parameters of the lattice structure of the parts, which reduced the manufacturing cost by 53.7% and the production time by 54.3% and increased the output. Arisoy et al. [27] used the surface response regression method to study the influence of the cooling rate and thermal gradient during laser processing of nickel alloy (IN625) material, which improved production efficiency.

In this study, TPU was used as the flexible polymer matrix material and CNTs as the conductive filler to print CPC-based pressure-sensing elements using SLS. First, CNT powder was dispersed in TPU powder using a strong acid oxidation method combined with a ball mill dispersion method, and the dispersion of the CNT powder was observed by combining with scanning electronic microscopy (SEM). A three-factor four-level orthogonal experimental design was performed, and the influence of the SLS process on the molding quality and sensing performance of the TPU/CNT sensing element was discussed. The optimized TPU/CNT sensing element was tested for its sensing performance (such as piezoresistive sensitivity, pressure measurement range, response time, and stability under cyclic load).

2 Materials and methods

2.1 SLS experimental equipment

Fabrication of the TPU/CNT sensing element and molding processing were mainly achieved using SLS equipment (AFS-360, Beijing Long Yuan Technology Ltd.) equipped with a CO_2 laser generator (laser power of 55 W and laser spot diameter of 0.20 mm). The dimensions of the forming box were 360 mm $\times$ 360 mm $\times$ 500 mm, with a layer thickness range of 0.1 to 0.3 mm.

2.2 Materials

Carbon nanotube powder with an average diameter of 5–15 nm and an average length of 0.5–1.5 μm was supplied by Shenzhen Turing Evolution Technology Co., Ltd., China. Thermoplastic polyurethane powder with an average grain diameter of 80 μm (LUVOSINT X92A-1) was purchased from Luvocom, Germany, as shown in Fig. 1a. Concentrated sulfuric acid (H_2SO_4 , 96–98%) and concentrated nitric acid (HNO_3 , 68%) were supplied by Sinopharm Chemical Reagent Co., Ltd., China.

Fig. 1 **a** Micromorphology of the TPU powder. **b** Micromorphology of the TPU/CNT powder

2.3 Preparation of TPU/CNT composites

The properties of the TPU/CNT sensing elements depend on the dispersion of the CNTs in the TPU matrix and on the interfacial binding force between the CNTs and TPU. Therefore, they were pre-treated using a strong acid oxidation method to directly modify the surface of the CNTs and improve their dispersion in the TPU matrix [28].

The TPU/CNT composites were prepared as follows: mix concentrated H_2SO_4 and HNO_3 in a beaker at a ratio of 3:1 with CNTs and magnetically stir them at 80 °C for 2 h. After standing and stratification, the CNTs were filtered out and washed with deionized water until the pH was neutral. Then, the CNTs were dried and put into a planetary ball mill with a grinding speed of 240 r/min and a grinding time of 2 h. The crushed CNT powder and TPU were put into an SHR50A high-speed mixer at a ratio of 0.25 wt% and stirred for 2 min at a speed of 700 r/min to obtain TPU/CNT composite powder. As shown in Fig. 1b, the CNTs are still slightly aggregated after strong acid oxidation and crushing, but they are evenly distributed on the surface of the TPU.

2.4 Methods

2.4.1 Orthogonal experiment design

The piezoresistive sensitivity, density, and dimensional precision of TPU/CNT sensing elements are important factors to evaluate sensing performance and formability. These factors mainly depend on main SLS process parameters such as laser power, layer thickness, and scan spacing. To determine the range of processing parameters, we conducted some experiments in advance. The experiments showed that both large layer thickness (0.3 mm) and low laser power (17 W) led to delamination of the part (Fig. 2a), and no resistance change can be detected. Furthermore, due to the limitation of the SLS equipment's accuracy, the minimum layer thickness was specified as 0.1 mm. High laser power (30 W) caused excessive sintering, resulting in warping and degrading of parts (Fig. 2b).

Hence, the laser power was set at a range of 19–28 W and the layer thickness range was 0.1–0.25 mm. Additionally, since the laser is a Gaussian heat source, the scan spacing has a great influence on the heat absorption of the material. When the scan spacing is large, a continuous sintered surface cannot be formed between the scanning lines. If the scan spacing is too small, the sintered powder will absorb too much heat, and the scanning lines will overlap considerably, resulting in excessive sintering. The sintering efficiency and quality will be unsatisfactory, so the scan spacing was set at a range of 0.1–0.25 mm. An SLS experiment was performed using an orthogonal experiment design scheme of three factors and four levels. The test scheme is shown in Table 1.

2.4.2 Scanning electronic microscopy

Scanning electronic microscopy (FEI Quanta 200 microscope, Hewlett-Packard Company, Netherlands) was used to observe the morphologies of the TPU/CNT powders and TPU/CNT sensing elements. Since the materials are non-conductive, the specimens were first sputtered with gold using SEM specimen-coating equipment.

2.4.3 Piezoresistive response tests

The piezoresistive response experiment equipment was mainly composed of two parts: a mechanical testing machine and a high-precision multimeter, as shown in Fig. 3. The electronic universal testing machine (Byes-3003, Bangyi Precision Meter Co., Ltd.) was used to provide external pressure, and the high-precision multimeter (Victor 8165A) was used to record the resistance change of the TPU/CNT sensing elements under pressure. The upper and lower sides of the SLS TPU/CNT sensing element samples ($\Phi 25 \times 3 \text{ mm}^3$) were glued with a pair of copper electrodes with conductive silver glue to ensure good contact between the samples and the electrodes. The sensing elements were compressed by jogging, and a high-precision multimeter was used to record changes in their resistance.

Fig. 2 TPU/CNT parts. **a** Layer-separation part. **b** Warping part

2.4.4 Piezoresistive sensitivity

The piezoresistive sensitivity of the TPU/CNT sensing elements can be expressed by the following formula [29]:

$$S = \frac{(\Delta R/R_0)\%}{\Delta P} \quad (1)$$

$$\Delta R = |R - R_0| \quad (2)$$

where ΔP is the relative loaded compressive force, R_0 is the initial resistance, and R is the resistance. To allow the TPU/CNT sensor element to have a good resolution under high-pressure loading, the sensitivity at 150 kPa was considered to be one of the evaluation indicators of the orthogonal experiment.

2.4.5 Density

The mass and dimensions of the TPU/CNT sensing elements were measured with an analytical balance and a vernier caliper. Herein, the density ρ is calculated by Eq. 3:

$$\rho = W/(l \cdot b \cdot h) \quad (3)$$

where W denotes the mass of sensing elements (g), l represents the length of sensing elements (mm), b is the width of sensing elements (mm), and h denotes the thickness of sensing elements (mm).

2.4.6 Dimensional precision

A rectangular specimen with dimensions of 20 mm × 20 mm × 6 mm was used in the precision analysis. The actual

dimensions of the TPU/CNT sensing elements were measured using a vernier caliper. Herein the dimensional precision is calculated via Eq. 4:

$$\delta (\%) = \left(1 - \frac{L_0 - L}{L_0} \right) \quad (4)$$

where δ represents the dimensional precision of sensing elements (%), L_0 denotes the standard dimension of sensing elements (mm), and L indicates the actual dimension of sensing elements (mm). The dimension of TPU/CNT sensing elements in the Z direction (direction of thickness) was also measured, and the Z -dimensional precision δ_z was calculated (%).

3 Results and discussion

3.1 Single-factor analysis

The piezoresistive sensitivity, Z -dimensional precision, and density of TPU/CNT sensing elements were used to evaluate the effect of SLS processes on the sensing performance and molding quality of TPU/CNT sensing elements, as shown in Table 2. The order of the range value of the piezoresistive sensitivity of TPU/CNT sensing elements from large to small was as follows: laser power > layer thickness > scan spacing. These results illustrated that the laser power had the greatest effect on piezoresistive sensitivity, while scan spacing had the least effect. Layer thickness had the largest effect on density, while laser power had the least influence, and scan spacing had the greatest influence on Z -dimensional precision, while laser power had the least influence.

Table 1 Factors and levels of laser sintering test

Levels	Laser power (W), [A]	Scan spacing (mm), [B]	Layer thickness (mm), [C]
1	19	0.10	0.10
2	22	0.15	0.15
3	25	0.20	0.20
4	28	0.25	0.25

Fig. 3 A schematic diagram of the test setup

3.1.1 Analysis of the effect of process parameters on piezoresistive sensitivity

Through the orthogonal experiment, the effects of the SLS process on the piezoresistive sensitivity of TPU/CNT sensing elements were obtained, as shown in Fig. 4. Figure 4a shows that with increasing laser power, the sensitivity of TPU/CNT sensing elements was increased. Owing to low laser power, the internal pores of the TPU/CNT sensing elements were too large, and the distance between the CNTs distributed in the TPU particles was too wide (Fig. 5a, b). It was not easy to form a conductive path. As the pressure increased, the number of conductive paths to be constructed was less than the number to be destroyed, resulting in smaller changes in piezoresistive sensitivity. As the laser power increased, the TPU particles inside the sensing element melted more (Fig. 5c), the conductive particle spacing was reduced, and it was easier to form a conductive path during loading. Additionally, it can be seen from Fig. 5c and d that when the laser power was 25 W, the inside of the sensing element was already dense, and when the power was increased to 28 W, spherical holes appeared inside the sensing element. This was because higher power causes the materials to overburn, and the generated gas could not be discharged and formed smaller closed holes, which led to a continued increase in sensitivity. Research by scholars such as Chen et al. [10] and Jung et al. [30] has shown that the micropores and porous structure may,

to some extent, increase the sensitivity of sensing elements, which also confirms the phenomenon produced in this study.

Figure 4b shows that as the scan spacing increased, the sensitivity first decreased and then increased. For TPU/CNT sensing elements with different scan spacings, the difference in internal pores mainly occurred in every single layer of the sensors. Through the transverse deformation of the TPU particles, the pores of the sensing element were reduced and the sensitivity was improved. As shown in Fig. 6a, when the scan spacing was small (0.10 mm), the internal pore spacing was small, and conductive paths were easily formed under pressure. The pores in the single layer became larger with an increase of the scan spacing (0.15–0.20 mm), and the distance caused by the increase of the scan spacing was longer, and the damaged conductive path was larger than the constructed conductive path, resulting in a decrease in sensitivity. Since the laser spot diameter is 0.20 mm, when the scanning distance increased to 0.25 mm, the powder inside the TPU/CNT sensing element was not completely fused, and there were fewer conductive paths formed in the lateral direction. As the pressure increased, the TPU particles were deformed laterally, the distance between the conductive particles was shortened, more conductive paths were constructed, and the sensitivity increased.

Figure 4c shows that as the layer thickness increased, the sensing element sensitivity first increased and then decreased. When the layer thickness was 0.1 mm, the inside of the sensing element was dense (Fig. 6b), and the polyurethane

Table 2 Range values of the effect of process parameters on the quality of TPU/CNT sensing elements

Levels	Laser power (W), [A]	Scan spacing (mm), [B]	Layer thickness (mm), [C]
Sensitivity (%/kPa)	5.5	1.138	5.229
Density (g/cm ³)	0.102	0.173	0.219
Z-dimensional precision (%)	2.188	8.646	4.438

Fig. 4 Effects of process parameters on the piezoresistive sensitivity of TPU/CNT sensing elements. **a** Laser power. **b** Scan spacing. **c** Layer thickness

particles moved less under applied pressure. The transition was mainly caused by the change of the internal conduction particle spacing, and the resistance change was not obvious, so the sensitivity was small. As the thickness of the layer increased, pores were generated inside (Fig. 6c). The possibility of contact between internal conductive particles gradually increased under applied pressure, thus increasing sensitivity. However, when the layer thickness increased to 0.25 mm, it was difficult to form a conductive path. When pressure was applied, the number of conductive paths constructed was smaller than the number destroyed, resulting in smaller changes in piezoresistive sensitivity.

3.1.2 Analysis of the effects of process parameters on density

The effects of the process parameters on the density of TPU/CNT sensing elements are shown in Fig. 7. Figure 7a shows that with an increase in laser power, the laser energy absorbed by TPU/CNT powder increased, the powder was completely fused, and the quantities of the internal pores of TPU/CNT

sensing elements decreased. Thus, the density of TPU/CNT sensing elements increased. When the input power of the laser remained constant, Fig. 7b and c show that with the increase in scan spacing, the re-sintering zone was reduced, the power absorbed by the TPU/CNT powder gradually decreased, the powder was not fully fused, and the amounts of internal pores gradually increased. Thus, the densities of TPU/CNT sensing elements decreased. With increasing layer thickness, the power absorbed by the combination zone between layers decreased, and the number of internal pores of TPU/CNT sensing elements increased. Therefore, the density of sensing elements decreased.

3.1.3 Analysis of the effect of process parameters on Z-dimensional precision

The effects of the process parameters on the Z-dimensional precision of TPU/CNT sensing elements are shown in Fig. 8. Figure 8a–c show that with increased laser power, the power absorbed by TPU/CNT powder increased. The power diffused

Fig 5 Micromorphology of the cross sections of the TPU/CNT sensing element with different laser power values. **a** Laser power 19 W. **b** A magnified image of **a**. **c** Laser power 25 W. **d** Laser power 28 W

Fig 6 **a** Schematic diagram of the conductive path formed by TPU/CNT sensing elements with different scan spacings after compression. **b** Micromorphology of the cross sections of the TPU/CNT sensing element with a layer thickness of 0.10 mm. **c** Micromorphology of the cross sections of the TPU/CNT sensing element with a layer thickness of 0.15 mm

along the laser sintering path and the sintered depth was increased, which increased the dimension of TPU/CNT sensing elements in the Z direction, decreasing the Z-dimensional precision. Following an increase in scan spacing, the re-sintering zone was reduced, and the sintered depth decreased, resulting in increased Z-dimensional precision. When the input power of the laser remained constant, with increasing layer thickness, the sintered depth decreased, and thus, the Z-dimensional direction of TPU/CNT sensing elements decreased. Therefore, Z-dimensional precision improved.

3.2 Multi-index synthetic analysis

3.2.1 Synthetic weighted evaluation

Multi-index test results were evaluated using a synthetic weighted scoring method. The synthetic weighted scoring method is a method by which multi-index weights were determined based on the importance of each test index in the whole test. The piezoresistive sensitivity, density, and Z-dimensional precision test results were transformed into the single-index test results, and then the test scheme was optimized according to a single-index analysis method.

The piezoresistive sensitivity, density, and Z-dimensional precision were transformed into dimensionless forms. The transformations were performed following Eqs. 5–7:

$$Z_1 = (S - S_{\min}) / (S_{\max} - S_{\min}) \quad (5)$$

$$Z_2 = (\rho - \rho_{\min}) / (\rho_{\max} - \rho_{\min}) \quad (6)$$

$$Z_3 = (\delta - \delta_{\min}) / (\delta_{\max} - \delta_{\min}) \quad (7)$$

3.2.2 Synthetic weights of test index

Sensitivity affects the sensing performance and measurement range of the pressure-sensing element. When the measurement range is sufficient, the higher the sensitivity, the better the resolution of pressure detection. The density of TPU/CNT sensing elements was higher, indicating that the inner structure of the sensing element was denser and that the internal conductive network structure was not easily destroyed. The higher Z-dimensional precision revealed better forming precision. The test indexes were given weights based on the importance of the test indexes. In general, higher weights were given to indexes of more subjective importance.

Fig 7 Effect of process parameters on the density of TPU/CNT sensing elements. **a** Laser power. **b** Scan spacing. **c** Layer thickness

Fig. 8 Effect of process parameters on Z-dimensional precision of TPU/CNT sensing elements. **a** Laser power. **b** Scan spacing. **c** Layer thickness

The effect of piezoresistive sensitivity was the key consideration, and the density and Z-dimensional precision were secondary considerations. The weights of piezoresistive sensitivity, density, and Z-dimensional precision were $\lambda_1 = 0.5$, $\lambda_2 = 0.3$, and $\lambda_3 = 0.2$, respectively. Therefore, the synthetic weighted scoring values are shown by Eq. 8:

$$Z = \lambda_1 Z_1 + \lambda_2 Z_2 + \lambda_3 Z_3 \quad (8)$$

3.2.3 Results and analysis of the test

Test data were processed using the synthetic weighted scoring method. The range analysis results of piezoresistive

sensitivity, density, and Z-dimensional precision were obtained, as shown in Table 3.

Scan spacing and layer thickness were large because the laser power of the fourth group of orthogonal experiments was too small, resulting in a slight layering phenomenon in the laser sintering sensing elements. Hence, so the resistance signal could not be detected, and the sensitivity was set to 0. Table 3 shows that the range of laser power (A) was the largest, which had the greatest effect on the test index. Level 4 was suitable for A. The range of layer thickness (C) was smaller than that of A, implying its effect was less. Level 3 was suitable for C. The range of scan spacing (B) was the smallest; hence, its influence was the least. Level 2 was suitable for C.

Table 3 Laser sintering process scheme and experimental results of TPU/CNT sensing elements

Serial number	Laser power (W) [A]	Scan spacing (mm) [B]	Layer thickness (mm) [C]	Test index			
				Sensitivity (%/kPa)	Density (g/cm ³)	Dimensional precision (%)	Synthetic weight
1	19	0.1	0.1	2.378	1.018	86.33	0.353
2	19	0.15	0.15	5.192	0.893	94.94	0.482
3	19	0.2	0.2	2.811	0.725	97.94	0.330
4	19	0.25	0.25	0	0.631	99.83	0.200
5	22	0.1	0.15	5.405	0.989	87.89	0.454
6	22	0.15	0.1	8	0.875	94.56	0.554
7	22	0.2	0.25	11.243	0.733	97.92	0.607
8	22	0.25	0.2	2.605	0.902	96.00	0.421
9	25	0.1	0.2	6.706	0.941	90.00	0.492
10	25	0.15	0.25	7.351	0.834	96.28	0.529
11	25	0.2	0.1	5.4	1.012	94.78	0.572
12	25	0.25	0.15	7.783	0.824	96.94	0.546
13	28	0.1	0.25	12.325	0.910	90.22	0.654
14	28	0.15	0.1	1.73	1.050	89.39	0.401
15	28	0.2	0.15	2.815	0.904	94.44	0.406
16	28	0.25	0.2	15.51	0.812	96.25	0.776
K1	0.341	0.488	0.437	Optimum combination: A4C3B2			
K2	0.509	0.492	0.472				
K3	0.535	0.479	0.538				
K4	0.559	0.486	0.498				
R	0.218	0.013	0.101				

Table 4 Contrast data of test results

Test contrast	Low loading		High loading		Density (g/cm ³)	Z-dimensional precision (%)
	Pressure (kPa)	Sensitivity (/kPa)	Pressure (kPa)	Sensitivity (/kPa)		
Optimized	3–70	1.357	70–240	0.0328	0.916	94.38
Optimum	30–70	0.991	70–240	0.162	0.812	96.25

Thus, A4C3B2 was the optimum combination of various factors, namely, the laser power was 28 W, the scan spacing was 0.15 mm, and the layer thickness was 0.2 mm.

The optimized process parameters were compared with the optimum combination of comprehensive weighted scores (Table 3), and the analysis results are presented in Table 4. Table 4 shows that using the optimized SLS process parameters, the density of the TPU/CNT sensing elements was 12.8% higher than that of the optimum combination, and the Z-dimensional precision was reduced by 1.94%. To further understand the changes of the optimum process parameters and the optimized sensing performance of the TPU/CNT sensing elements, the piezoresistive sensitivity of the TPU/CNT sensing elements are plotted in Fig. 9a for comparison. Figure 9a shows that when high loading was applied, although the TPU/CNT sensing elements prepared with the optimum process parameters were more sensitive than the optimized sensing element, they were more sensitive to detecting large pressure loads. However, due to large internal pores in the TPU/CNT sensors, no internal conductive network was formed in the initial stage of pressure loading, and the resistance signal

could not be detected until the load reached 30 kPa. Additionally, during the pressure unloading process, the optimum parameter sensing elements had a certain hysteresis, and when unloaded to 40 kPa, the resistance could not be detected when unloaded to 40 kPa. It may be because there were many pores in the TPU/CNT sensing elements. When high loading is applied, the internal conductive network structure was destroyed and could not be restored to its initial resistance.

A wide pressure detection range (3–240 kPa) and no obvious hysteresis during pressure loading and unloading were seen because the optimized TPU/CNT sensing elements could detect small pressures. Although sensitivity was low under large pressure loads, it was almost the same as that of the flexible pressure-sensing elements reported by He et al. [31] and Yang et al. [32]; thus, the optimized TPU/CNT sensing elements were used for subsequent research on their sensing performance.

In addition to sensitivity and pressure detection range, the response time and reproducibility during cyclic testing were also important indicators for identifying important sensors. Therefore, we conducted a follow-up experimental test on

Fig. 9 **a** Comparison of resistance change ($\Delta R/R_0$) curves of the optimized TPU/CNT sensing elements and optimum TPU/CNT sensing elements. **b** The resistance changes after a sudden increase in pressure from 22 to 30 kPa. **c** Long-term stability of the TPU/CNT sensing elements over 500 cycles at a compression strain of 10%

Table 5 Contrast data of test results

Materials	Sensitivity: /kPa	Pressure range: kPa	Response time: ms	Processing technology	Ref. number
TPU/CNT composites	0.162–0.991	3–240	33	SLS technology	This manuscript
MWCNT and TPU foam	0.013–0.032	0–200	29	Direct attachment method	[15]
MWCNT/PU composites	0.0428	0–63	–	Casting technology	[31]
Graphene foam	0.36	0–5	80	Chemical and thermal reduction processes	[32]
MWCNT and rGO loaded PU foam	0.034–0.088	0–48.8	30	Drying directly after soaking	[33]

the sensing performance of the optimized TPU/CNT sensing elements. As shown in Fig. 9b, the response time to a sudden pressure input of 30 kPa was 33 ms. Compared with the response times of other sensors, this response time was almost identical to the value from other sensors [15, 33]. Meanwhile, the durability and stability of the TPU/CNT sensing elements were further evaluated over 500 cycles at a compression strain of 10% (Fig. 9c). The resistance changed periodically, and no obvious fluctuation of the sensing signal was observed. After 500 compression cycles, the strain sensing pattern remained stable, and the repeatability error was less than 3.58%, indicating that the conductive network was very stable, which was beneficial for stable sensing. All of these indicate that the optimized TPU/CNT sensing elements have high repeatability, fast response, stability, and durability. To compare the optimized TPU/CNT sensing elements with flexible pressure

sensors prepared by other processing technologies, the comparison report of the main parameters of the latest pressure sensor is presented in Table 5. The report shows that the SLS TPU/CNT sensor has the advantages of high sensitivity, good stability, and fast response time, but with a small pressure, it has no resistance response. In a future work, we will continue to explore post-processing and broaden the pressure measurement range.

3.3 Application

To illustrate the applicability of the sensing performance of TPU/CNT sensing elements, we used a three-dimensional scanner to scan and model the human foot and used laser-sintered TPU/CNT composites to prepare pressure-sensing insoles to detect the distribution of plantar pressure (Fig. 10a). The upper

Fig. 10 **a** The TPU/CNT sensing insole preparation process. **b** Resistance change ($\Delta R/R_0$) of four sensing positions under single-point pressure-loading experiment. **c** Fitting values of pressure distribution and resistance changes at four sensing positions of the sole

and lower sides of the four main positions of the sensing insole were, respectively, bonded with a pair of copper electrodes and conductive silver glue to ensure good contact between the insole and the electrodes. The four groups of electrodes were connected to a high-precision multimeter to collect the resistance signal of the plantar pressure. The four pressure-sensing positions were the first metatarsal bone, the second and third metatarsal bones, the fourth and fifth metatarsal bones, and the heel (Fig. 10a).

Before the test, the pressure-sensing insole was placed on the pressure-loading platform, and the single-point pressure-loading experiment was carried out on the four test points. To calibrate the pressure load of each test point, control the change of the loading force through the controller and record the resistance change of each sensing position. During the test, the subject took off his shoes, with feet shoulder-width apart and stood on the sensing insole. The test time was 9–10 s, and the test was performed five times, and the results were inputted into a computer; the data were fitted to measure the pressure experienced by each test point.

Figure 9b shows the resistance changes of the four pressure detection positions. Since the sensing insoles were modeled according to the human foot, the generated complex three-dimensional curved surface had a slight impact on the resistance changes of each test point (Fig. 10b). Figure 10c shows the fitting values of pressure distribution and resistance changes at four sensing positions of the insole. The highest pressure was detected at the fourth and fifth metatarsal bones, at approximately 191.22 kPa, followed by the second and third metatarsal bones and the heel, and the first metatarsal bone experienced the least stress. The detected pressure range was consistent with the normal adult plantar pressure range reported by Zhang [34] and Veves et al. [35]. Therefore, it has been demonstrated that the optimized TPU/CNT sensing elements can be used to measure biological and motion signals, and sensing elements can be installed according to demand, providing new technical solutions for human movement detection, smart wearable devices, medical rehabilitation, and other fields.

4 Conclusions

The orthogonal experiment design method and the comprehensive weight method were used to optimize the multi-objectives and prepare the TPU/CNT pressure sensor element with good flexible sensing performance. The optimized SLS TPU/CNT sensing element density was increased by 12.8%; the Z-direction accuracy was reduced by 1.94%, and the sensing element has a wide pressure detection range (3–240 kPa) and high sensitivity (1.357/kPa for 3–70 kPa, 0.0328/kPa for 70–240 kPa); its response time can reach 33 ms, and it still has good stability and durability after 500 loads (10% strain).

Finally, the flexible pressure-sensing element was successfully applied to detect human foot pressure. This work provides a good research foundation for smart wearable devices and human motion detection and presents a simple and effective method to develop flexible pressure sensors.

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Compliance with ethical standards

Conflict of interest The authors declare that they have no conflict of interest.

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