Transparent and flexible force sensor based on microextrusion 3D printing

Runhuai Yang[1,], Tianyun Gao[1,*], Didi Li[1], Yueming Chen[1] ✉, Guoqing Jin[2], Haiyi Liang[3,4], Fuzhou Niu[5]*

[1]*Department of Biomedical Engineering, School of Life Science, Anhui Medical University, 230032 Hefei, People's Republic of China*
[2]*Robotics and Microsystems Center, School of Mechanical and Electric Engineering, Soochow University, 215006 Suzhou, People's Republic of China*
[3]*CAS Key Laboratory of Mechanical Behavior and Design of Materials, University of Science and Technology of China, 230027 Hefei, People's Republic of China*
[4]*IAT-Chungu Joint Laboratory for Additive Manufacturing, Institute of Advanced Technology, University of Science and Technology of China, 230026 Hefei, People's Republic of China*
[5]*College of Mechanical Engineering, Suzhou University of Science and Technology, 215000 Suzhou, People's Republic of China*
[*]*Runhuai Yang and Tianyun Gao contributed equally to this work.*
✉ *E-mail: ahmubmechen@163.com*

Published in Micro & Nano Letters; Received on 30th October 2017; Revised on 12th June 2018; Accepted on 21st June 2018

Transparent and flexible force sensor provides potential applications in a lot of area such as human wearable sensor, robotic sensor and underwater equipment. Here the work presents a zig-zag patterned hydrogel-based force sensor. Slow-gelling alginate hydrogel, self-designed microextrusion 3D printer and polydimethylsiloxane (PDMS) were adapted for the sensor fabrication. A slow-gelling hydrogel printing ink was prepared and printed onto the treated PDMS layer with a computer-aided designed zig-zag pattern. The line width and resistance of the printed hydrogel were precisely controlled by the self-designed microextrusion 3D printer. To stabilise the water content, a sandwich structure was adopted and two PDMS cover layers were fabricated to seal the hydrogel sensor. Theoretical analysis was performed and it is shown that the change rate of resistance was linear related to the force, and this analysis was proved by experiment. Experiments also show that the sensor was flexible and transparent. The zig-zag patterned hydrogel also and the PDMS layers provided stable water content and recording.

1. Introduction: The study of flexible sensors has grown significantly. High-sensitivity, soft and stretchable and transparent sensing mechanisms play critical roles in applications such as wearable equipment, artificial skin and the sensors for soft robotics [1]. Although many flexible sensors exist, including liquid metals, nanofibrous, nanowires etc. [2], most of these materials have limited mechanical properties and low optical transparency [3]. Besides, the biocapability of these materials is also limited. Therefore, the applications of such sensors for long-term integrating with human being are limited. For example, flexible force sensor can be used on patient skin for the recovery of skin sensory capability of some skin damaged patients. These patients require a touch-neuron stimulus training, while the touch sensor provides a tactile sensation and a stimulus for neuron is performed simultaneously [4]. However, conventional flexible sensors yield potential harm such as metal leakage [5] and hard–soft material interfacial failure [6]. The low transparency also limited the integration with light control for the substrate.

The improvement of the stretchability, transparency and biocapability of the flexible sensors will extend their application scope. Recently, it is reported that the hydrogel was adopted as a wearable equipment which can measure the strain and finger bending [7] or measure the touch position [8]. These structures used the hydrogel as the top layer on the sensor. However, it is well known that the hydrogel keeps loss water in the air, and the resistance of the sensor could be strongly affected by the water content of the hydrogel [9]. The change of water content could affect the measure accuracy, and our experiments also show that in room temperature and common humidity (40–70%), the water content of hydrogel keeps decreasing. Therefore, the sensor should maintain the water content in service environment.

Compared to a straight line, the main advantage of zig-zag pattern is that it induces an increase of its electrical resistance [10]. Resistance is a critical parameter to the touch sensor. While a low resistance could induce a high current and increase the temperature which could reduce the measurement accuracy. Therefore, a zig-zag pattern can improve the stability of the sensors. Besides, the area of zig-zag pattern is easily controlled and the measurement area can be regulated. To print the hydrogel into a certain pattern, a 3D printer should be designed for hydrogels. Recently, most of the alginate hydrogel was mould by immersing the hydrogel structure in the solution of calcium chloride [11]. However, the mould of alginate hydrogel by immersing in calcium chloride causes the inhomogeneous of the hydrogel structure [12]. While the inhomogeneous of the hydrogel structure may reduce the precision of the sensor, a slow-gelling alginate hydrogel provides homogeneous distribution of the printed hydrogel and improves the measurement of sensor.

Therefore, a flexible force sensor was proposed based on a self-designed microextrusion 3D printing method and slow-gelling alginate hydrogel in this Letter. To improve the measurement of the sensor, the slow-gelling hydrogel was printed in a zig-zag pattern of parallel lines with width on the order of hundreds of micrometre. To increase the stability of the sensor, a sandwich structure was proposed to maintain the water content of the hydrogel in the sensor. The experimental results show that the sensor was transparent and flexible. The stability of water contents and the sensitivity of the sensors were improved, show that our design of the sensor was effective.

2. Materials and methods: Before the hydrogel was printed, a polydimethylsiloxane (PDMS) substrate was prepared firstly. The PDMS and curing agent (Sylgard 184 silicone elastomer, Dow Corning) were used as the substrate and the cover of the sensor. The PDMS was used in the recommended 10:1 ratio. After a 10 min mixing cycle in a planetary centrifugal mixer (Topoint, SPS-500) at 1500 RPM, it was spined on a quartz plate by a spin

doi: 10.1049/mnl.2018.5076

coater with 150 RPM for 3 min. Then the PDMS was moulding in 50°C by a heating platform for 2 h. Prior to hydrogel printing, the PDMS substrate was exposure to UV (200 W, 365 nm) for 2 h to improve the hydrophilia.

To obtain a homogeneous distribution of printed hydrogel, a slow-gelling alginate hydrogel was used. An inactivated form of calcium (Ca-Ethylenediaminetetraacetic acid (EDTA)) and a slowly hydrolysing molecule (glucono delta-lactone, GDL) was adopted to achieve this goal. The GDL induces the slow release of calcium ions from the Ca-EDTA. With 67.4 mM GDL concentrations, 20 min passed for gelation and >5 h was required to reach a plateau for the slow Ca^{2+} releasing Ca-EDTA/GDL method [13]. After that, the hydrogel pattern should be kept in a controlled temperature and humidity for 5 h to fix the printed structure. It is recommended that the printing could be finished in 20 min to avoid any failure or defect. The temperature and humidity were controlled by a chamber with constant temperature and humidity.

To print the slow-gelling alginate hydrogel into a certain pattern, a self-designed microextrusion 3D printer was built up, and it is shown in Fig. 1. Before the printing was start, the slow-gelling alginate hydrogel was loaded in the nozzle. A control unit was used to control the position and the pressure of the nozzle. A water circulation and a temperature control were used to control the temperature of the print platform. To ensure the hydrogel can be printed immediately after the slow-gelling hydrogel solution was mixed by the centrifugal mixer, the nozzle was designed as the container which could directly mixed in the mixer. By controlling the position of the nozzle and applying appropriate pressure, the slow-gelling hydrogel solution was printed into certain pattern. The temperature of the platform was also controlled to regulate the viscosity of the solution.

The fabrication and architecture of the force sensor are shown in Fig. 2. The microextrusion 3D printing was used to print the slow-gelling hydrogel in a zig-zag pattern. CAD software was used to print hydrogel structure [14]. Here, the hydrogel pattern was modelled by CAD software and was robotically deposited on the pre-treated PDMS substrate by a self-designed microextrusion printer. After the slow-gelling hydrogel was printed as zig-zag pattern, the sample was kept in a chamber with constant humidity (90%) and temperature (25°C) for 5 h. After the hydrogel was gelated, The PDMS solution was spined and mould on the sample. Consequently, a sandwich structure was fabricated by attaching two PDMS layers to a zig-zag patterned hydrogel layer as shown in Fig. 2*d*. To measure the resistance change of the sensor, an alternating voltage source was used to generate a 1 kHz and 0.5 V voltage and an alternating current meter was used to record the current, as shown in Fig. 2*d*.

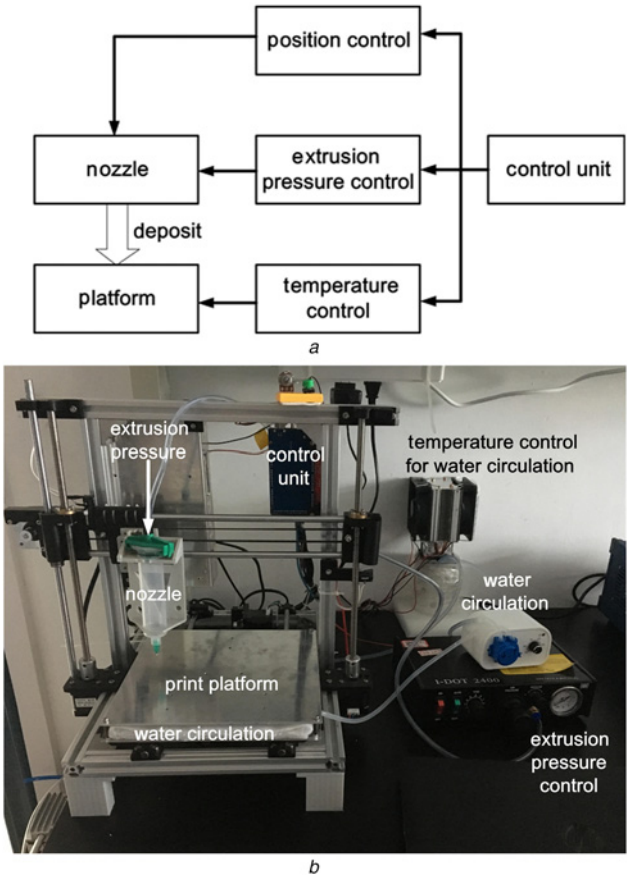

Fig. 1 *Structure of the microextrusion system*
a Illustration of the microextrusion 3D printing method
b Image of our self-designed microextrusion 3D printer

3. Results and discussions: Fig. 3 shows the process that the sensor has been designed and fabricated. CAD software was used

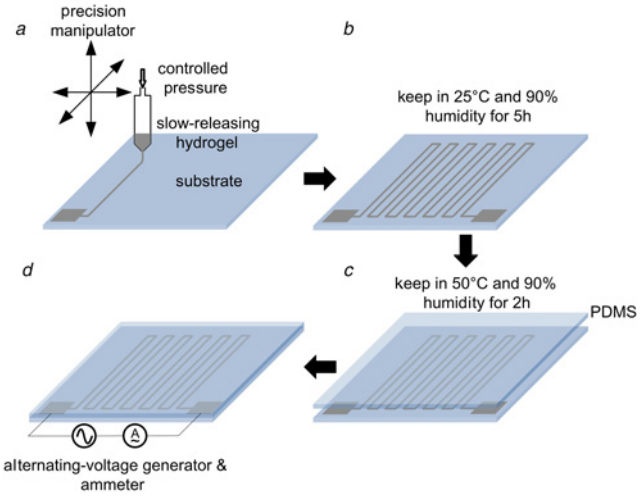

Fig. 2 *Schematic of the fabrication of the force sensor based on 3D printing*

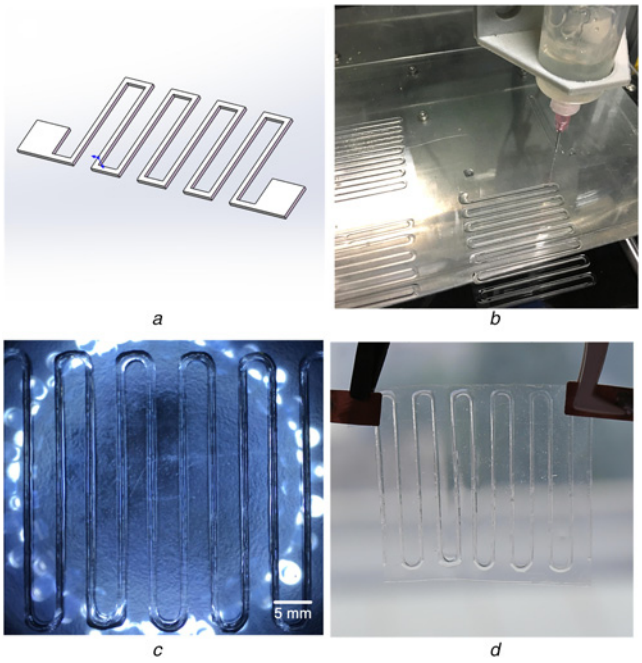

Fig. 3 *Zig-zag pattern*
a Designed in CAD software
b Printed by microextrusion printers
c Image shows that after printing, the hydrogel was mould without defect
d Finished sensor is flexible and transparent

to design the zig-zag pattern [14], as shown in Fig. 3*a*, the hydrogel was extrude by the self-designed microextrusion printers. Besides, more than one sensor can be printed by the 3D printing in one print period, as shown in Fig. 3*b*. This sensor has the potential capability to be high-throughput fabricated if a multiple nozzle 3D printer is adopted. Fig. 3*d* shows the finished sensor, which has a sandwich structure that the zig-zag patterned hydrogel was covered in two PDMS layers.

The theory of our sensor is based on the resistance change of hydrogel during a force is applied to the sensor. We assumed the cross section of the printed hydrogel line as semicircle, and the resistance of gel is calculated by (1), as follows:

$$R = \rho \frac{D}{\text{area}} = \rho \frac{D}{0.5\pi r^2}. \tag{1}$$

where ρ is the electrical resistivity of hydrogel, D is the total length of the printed zig-zag patterned hydrogel, and the r is the height of the hydrogel. While a force is applied to the sensor, a strain is happened. We assumed the resistivity is independent of stretch. Therefore, the change rate of resistance can be approximately calculated by total derivative in (2), as follows:

$$\mathrm{d}R \simeq \frac{\rho\, \mathrm{d}D}{0.5\pi r^2} - 2\frac{\rho\, D\, \mathrm{d}r}{0.5\pi r^3} \tag{2}$$

Divide both sides by R, and use (1), we have:

$$\frac{\mathrm{d}R}{R} \simeq \frac{\mathrm{d}D}{D} - 2\frac{\mathrm{d}r}{r} \tag{3}$$

Although the pattern of our sensor is zig-zag rather than straight line, we still can similarly consider the $\mathrm{d}D/D$ as the axial strain (on the printed long side) and the $\mathrm{d}r/r$ as the transverse strain, we use Poisson's ratio to further simplify the equation, and we have:

$$\frac{\mathrm{d}R}{R} \simeq (1 + 2\mu)\frac{\mathrm{d}D}{D} \tag{4}$$

where μ is the Poisson's ratio of the sensor. Since Poisson's ratio of PDMS is nearly 0.5, and Poisson's ratio of hydrogel can be considered as same as incompressible water, 0.5, therefore, from (4) we can know that change rate of the resistance keep a linear relation with the strain while the strain is not significant. While the strain also keep a linear relation with stress in the elastic range, we can assume that the change rate of resistance keep a linear relation with the force applied in a certain area on the sensor in the elastic range of the sensor.

The air pressure to extrude the hydrogel and the nozzle diameter (φ) is the key parameters which determine the extrusion output. The extrusion under different air pressure and different the inner-diameter (φ) of the microextrusion head was investigated. If $\varphi < 0.26$ mm and the pressure < 0.5 MPa, the hydrogel cannot be continuously pushed out. Fig. 4*b* shows the continuously printed hydrogel line width, and Fig. 4*c* shows the influence of the air pressure and φ on line width. With the increase of φ, the line width increased. The line width was more than the nozzle diameter, because of expansion after the extrusion. With the increase of air pressure, the line width also increased and they kept a nearly linear relationship. Compared to the recent results of hydrogel-based sensor [7], a finer hydrogel width (0.25 mm compared to 0.8 mm) can be obtained. Therefore, a delicate pattern can be printed by our hydrogel line. Another benefit by controlling the line width is that the resistance of the sensor was precisely regulated, as shown in Fig. 4*d*. A range of resistance from 13,000 to 350 Ω was obtained. This range shows that the sensor has a wide application field, for example 350 and 1000 Ω is a typical value of commercial strain gauge, and the sensor can be easily fabricated

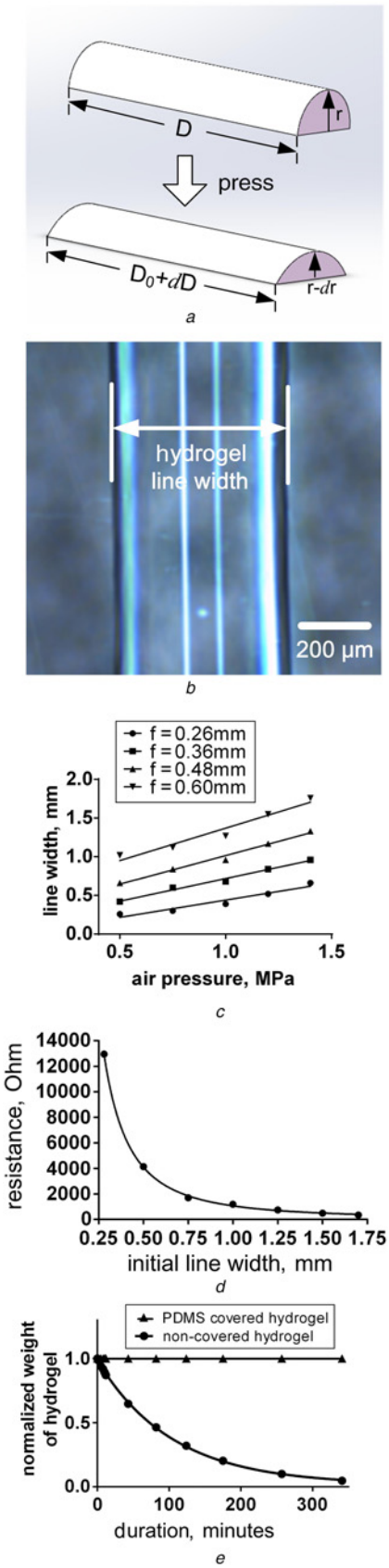

Fig. 4 *Study of the hydrogel part of the sensor*
a Illumination of the shape change of the sensor while a force is applied on it
b Measurement of the printed hydrogel line
c Influence of the nozzle diameter (φ) and the air pressure to the hydrogel diameter
d Influence of the line width to the electrical resistance of the sensor
e Recording of the hydrogel weight in room temperature (25°C) and common humidity (60%)

doi: 10.1049/mnl.2018.5076

with existing commercial strain gauge system. Besides, if the situation requires the sensor works with quite low current, a high resistance with thin line width can be used.

The water content changed in room temperature and humidity was recorded, as shown in Fig. 4*e*. Same as previous structure of hydrogel sensors [8], the hydrogel which was directly exposed to the air was also fabricated and the water content was studied. The change of the hydrogel weight was shown as the 'non-covered hydrogel'. It is obviously that the non-covered hydrogel lost water quickly, while the weight of hydrogel with PDMS covers almost unchanged in the whole duration. This comparison proved that our sandwich structure provides a stable water content for the sensor to have a stable recording.

A fabricated hydrogel sensor was tested by force in order to ensure that the force can be recorded by the change of resistance, as shown in Fig. 5. A universal testing machine was used to apply a compression force on the force sensor. The influence of the force on the resistance was shown in Fig. 5*a*, where R_0 presents the initial resistance of the hydrogel sensor while 0 N force was applied. ΔR presents the change of the resistance. Therefore, the ΔR divided by R presents the relative change rate of the resistance

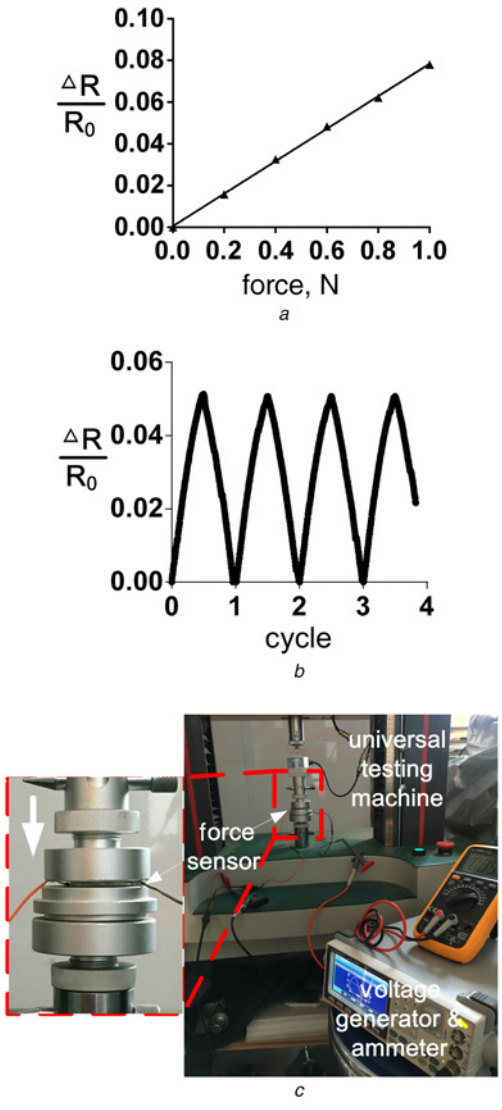

Fig. 5 *Force test experiments*
a Change of resistance induced by different force for both zig-zag patterned sensor and rectangle sensor
b Recycled experiments (1 N, 0.15 mm/s) of the zig-zag patterned sensor
c Force experimental setup

while force was applied. As can see from Fig. 5*a*, with the increase of the force, the relative change rate of the resistance kept a nearly linear increase. Such an experimental result with linear relation agrees well with the theoretical prediction. The cycle experiments that a 1 N force was cyclic loaded was also studied, as shown in Fig. 5*b*. The duration for each cycle is 2 s and the speed of the experiment is 0.15 mm/s. The result shows that the hydrogel-based force sensor had a stable response to the cyclic loading. Benefit from the sandwich structure and the slow-gelling hydrogel, the water contents of the hydrogel part kept stable and homogeneity. These features may provide advantages to the sensor so that it can have a stable force recording during cyclic loadings.

After we obtained a transparent, flexible and biocompatible force sensor, one of the future works regarding the sensor is to recovery the skin sensory capability of some skin damaged patients. These patients require a long-term touch-neuron stimulus training, while the touch sensor provides a tactile sensation and a stimulus for neuron is performed simultaneously [4]. All the materials we used are biocompatible and have no toxic or injurious effects on biological systems. Therefore, compared to other force sensors which yield potential harm such as liquid metal leakage, this sensor has the ability that it can interact with skin for a long time without any harm. Another advantage of our sensor is that it can integrate with optogenetic to obtain the touch-neuron stimulus training. Optogenetic technique provides a simple and efficient way to non-invasively stimulate peripheral neurons by light [15]. Therefore, transparent is necessary so that the control light can illuminate and stimulate the neuron through the sensor while the sensor is sensing the tactile on the skin right above the neuron. Such a soft and biocompatible touch sensors will allow human beings to comfortably use wearable devices during intimate interactions especially when it was interacting with children [16]. Benefit from biocompatibility, it also has the potential application to be an implantable sensor which can measure force inside human body. Besides, transparent property of this sensor may also provide the ability that it can be fabricated on a military robotics, while the supporting substrates have a cheating changeable colour.

4. Conclusion: In summary, we have presented a transparent and flexible force sensor based on self-designed microextrusion 3D printing and slow-gelling hydrogel. Our theoretical and experimental results demonstrated that our fabrication processes provide a simple way to obtain a hydrogel sensor with stable water content and high sensitivity, and the output signals were also repeatable and stable. In future work, the print resolution can be further improved and the sensor can be smaller, and the sensor will be fabricated as a wearable sensor for the recovery of the skin sensory capability of some skin damaged patients, and also be fabricated as an artificial skin for robotics.

5. Acknowledgments: This work was supported by the National Natural Science Foundation of China (grant nos. 61603002 and 61773274), the Scientific Research of BSKY (grant no. XJ201517) from Anhui Medical University, the Plan of Funding Outstanding Innovation Projects Launched by Talents Returning from Studying Overseas of Anhui Province.

6 References

[1] Gong S., Schwalb W., Wang Y., *ET AL.*: 'A wearable and highly sensitive pressure sensor with ultrathin gold nanowires', *Nat. Commun.*, 2014, **5**, pp. 1–8
[2] Ponce Wong R.D. Posner J.D., Santos V.J.: 'Flexible microfluidic normal force sensor skin for tactile feedback', *Sens. Actuat. A Phys.*, 2012, **179**, pp. 62–69
[3] Novak D., Riener R.: 'A survey of sensor fusion methods in wearable robotics', *Robot. Auton. Syst.*, 2015, **73**, pp. 155–170
[4] Chortos A., Liu J., Bao Z.: 'Pursuing prosthetic electronic skin', *Nature Mater.*, 2016, **15**, (9), pp. 937–950

[5] Menguc Y., Park Y., Martinez-Villalpando E., *ET AL.*: 'Soft wearable motion sensing suit for lower limb biomechanics measurements'. Proc. IEEE Int. Conf. Robotics and Automation, Karlsruhe, Germany, 2013, pp. 5309–5316
[6] Suo Z.: 'Mechanics of stretchable electronics and soft machines', *MRS Bull.*, 2012, **37**, (3), pp. 218–225
[7] Tian K., Bae J., Bakarich E., *ET AL.*: '3D printing of transparent and conductive heterogeneous hydrogel–elastomer systems', *Adv. Mater.*, 2017, **29**, (10), pp. 1–8
[8] Kim C.-C., Lee H.-H., Oh K. H., *ET AL.*: 'Highly stretchable, transparent ionic touch panel', *Science (80-.)*, 2016, **353**, (6300), pp. 682–687
[9] Ahmed E.M.: 'Hydrogel: preparation, characterization, and applications: a review', *J. Adv. Res.*, 2015, **6**, (2), pp. 105–121
[10] Stassi S., Cauda V., Canavese G., *ET AL.*: 'Flexible tactile sensing based on piezoresistive composites: a review', *Sensors*, 2014, **14**, (3), pp. 5296–5332
[11] He Y., Yang F., Zhao H., *ET AL.*: 'Research on the printability of hydrogels in 3D bioprinting', *Sci. Rep.*, 2016, **6**, (1), p. 29977
[12] Kuo C.K., Ma P.X.: 'Ionically crosslinked alginate hydrogels as scaffolds for tissue engineering: part 1. Structure, gelation rate and mechanical properties', *Biomaterials*, 2001, **22**, (6), pp. 511–521
[13] Du G., Wu F., Cong Y., *ET AL.*: 'Versatile controlled ion release for synthesis of recoverable hybrid hydrogels with high stretchability and notch-insensitivity', *Chem. Commun.*, 2015, **51**, pp. 15534–15537
[14] Lee J.S., Hong J.M., Jung J.W., *ET AL.*: '3D printing of composite tissue with complex shape applied to ear regeneration', *Biofabrication*, 2014, **6**, (2), pp. 1–12
[15] Towne C., Montgomery K.L., Iyer S.M., *ET AL.*: 'Optogenetic control of targeted peripheral axons in freely moving animals', *PLoS One*, 2013, **8**, (8), pp. 1–10
[16] Wijk U., Carlsson I.: 'Forearm amputees' views of prosthesis use and sensory feedback', *J. Hand Ther.*, 2015, **28**, (3), pp. 269–278

doi: 10.1049/mnl.2018.5076