

Shojaei Baghini, M. and Dahiya, R. (2022) Direct Ink Writing of Tunnelling Graphite Based Soft Piezoresistive Pressure Sensors. In: 2022 IEEE International Conference on Flexible and Printable Sensors and Systems (FLEPS 2022), Vienna, Austria, 10-13 Jul 2022, ISBN 9781665442732 (doi: [10.1109/FLEPS53764.2022.9781527](https://doi.org/10.1109/FLEPS53764.2022.9781527))

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# Direct ink writing of tunnelling graphite based soft piezoresistive pressure sensors

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**Abstract**—Tunneling based piezoresistive sensors are often utilized for dynamic pressure sensing due to their low cost, ease of fabrication, ability to be printed and integrated with read-out modules. These devices can be subsequently integrated with transistors, actuators and other components towards the development of multifunctional electronic skin (e-Skin), where it is important that sensors exhibit uniform and replicable behavior. This can also help to minimize the need for compensation circuits during long-term use. In this study, direct ink writing of custom-made low viscosity graphite ink is used to develop soft piezoresistive pressure sensors. The uniformity of the devices is gauged via the base conductivity and piezoresistive response, both of which exhibit a very good coefficient of variation of 2.21% and 7.1%, respectively. Furthermore, the sensors are sensitive to a wide range of forces from 0-7 N (~3.2 MPa maximum pressure). These devices pave the way towards efficient integration of pressure sensors for object grasping and manipulation due to their small size and bendability.

**Keywords**—pressure sensor, tunneling sensors, piezoresistance, direct ink writing, flexible sensors

## I. INTRODUCTION

Accurate and repeatable pressure sensors play a significant role in the development of electronic skin (e-Skin) [1-3] for applications such as rehabilitation [4, 5] robotics [6, 7] interactive systems [8, 9] and health monitoring [10, 11]. Among a wide variety of transductions mechanisms explored for pressure sensing, the tunnelling based piezoresistive sensors provide an effective method towards improved tactile recording due to their low cost, high gauge factor, thermal stability and compatibility to the printing technology [12, 13]. Furthermore, sensitivity of these sensors can be carefully controlled via engineering the active materials along with their contacts. These sensors can be seamlessly integrated onto curvilinear surfaces, leading to improved conformability as well as spatial resolution. This makes them the ideal for the sensory feedback needed for grasp control and object manipulation [14-16]. Furthermore, hysteresis in the piezoresistance response curve can potentially be utilized to fabricate mechanical sensors with touch memory.

Soft piezoresistive pressures sensors are often developed with a variety of nanocomposites embedded in soft elastomers. Among these, the carbon based fillers have gained considerable traction due to their low costs, ability to be integrated with soft polymers and shear thinning solvents [17]. The latter aspect is significantly advantageous for the fabrication of sensors with improved repeatability with thickness as low as 100  $\mu\text{m}$  via direct ink writing (DIW). The shear thinning properties of the

fabricated inks lead to intrinsic orientation in the graphite particles i.e. the agglomerated particles align to the direction of shear during print thus improving material conductivity and enabling the printing at low pressures which does not lead to damage of flexible substrates [18]. The working principle of typical soft piezoresistive sensors is shown in Fig. 1. Under normal condition, the conductive particles in the soft elastomeric matrix are held together through binders. However, they form the conductive tunnelling pathways under the application of a force, which leads to a change in the resistance wherein the number density of captured charge carriers is function of electrode material and area. In terms of forces associated with the skin, grip forces for light and soft objects occur in the range 1-100 kPa which falls within the region of sensitivity of the filler based sensors [19, 20].

A wide variety of soft piezoresistive sensors have been developed using a mix of photolithography and printing (e.g. screen printing) steps [21, 22]. However, one-step patterning of soft sensors using method such as DIW is desired due to reduced complexity, resource efficiency and improved repeatability as process-related variables are significantly reduced. In this regard, precise printing can be utilized to fabricate sensors responsive to a range of tactile pressures corresponding to soft touch and larger grasping stresses required for object manipulation [23, 24]. The focus of this paper is on the fabrication of piezoresistive sensors via DIW for areas such as fingertips and investigate their repeatability in terms of conductivity and piezoresistive response. Whilst, direct ink writing based piezoresistive sensors have been widely reported, their replicability is not deeply investigated [25-27]. In order to achieve effective integration with other sensing and transducing modules such as transistors and actuators, the uniformity of the sensors must be analysed and taken into account.

This paper is organised as follows: Section II discusses the fabrication of the sensors using DIW, Section III details the

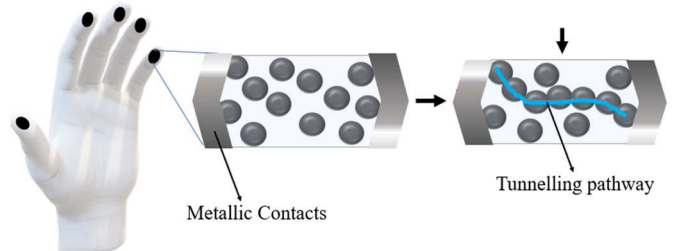


Fig. 1. Fingertip integrated sensors with tunnelling piezoresistivity on application of normal force.

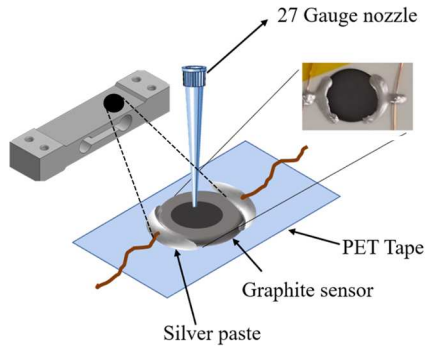


Fig. 2. Fabrication scheme of presented piezoresistive sensors [scale = 5 mm] along with schematic of beam type load cell used during characterization.

characterization of the sensors as well as the hysteresis response. A summary of the key results, conclusion and future aspects are provided in Section IV.

## II. MATERIALS AND METHODS

### A. Preparation of graphite ink for DIW

The graphite ink utilized in this study is prepared in-house based on a previous reported study [3]. The graphite content by weight percentage has been reduced in order to obtain ink with lower viscosity and shear thinning properties suitable for printing. The ink consists of terpineol (~95%, Sigma Aldrich) as the solvent, 2 w/w% graphite powder (<20  $\mu\text{m}$ , synthetic, Sigma Aldrich), 1 w/w % ethyl cellulose (48.0-49.5% (w/w) ethoxyl basis, Sigma Aldrich) as binder and 100  $\mu\text{l}$  of Triton X-100 (laboratory grade, Sigma Aldrich) as surfactant to improve dispersion stability. Once a uniform dispersion of ethyl cellulose and terpineol is achieved by magnetic stirring at room temperature, the graphite powder is added gradually and further stirring is carried out overnight. The graphite powder is grinded manually in a ceramic mortar prior to usage to improve the size distribution of the particles, contributing to better uniformity. We expect that this process contributes to breaking down larger graphite articles.

### B. DIW of tunneling piezoresistive sensors

Printing of the ink is carried out on a flat polyethylene terephthalate (PET) tape of thickness equal to 250  $\mu\text{m}$  which can be subsequently peeled off after printing and attached conformably to curvilinear surfaces. The uniformly dispersed conductive graphite ink is loaded in a syringe barrel with a 27 gauge (200  $\mu\text{m}$  in diameter) 30 mm long nozzle attached at the end. The printing of three sensors is carried out by using an automated 3D Bioprinter (Brinter®, Finland) at controlled temperature. Each sensor has a diameter of 8 mm with an active impact area of 19.6  $\text{mm}^2$ . The print was configured towards fabrication of thin single layered devices with an average thickness of 250  $\mu\text{m}$  measured using profilometry (DektakXT Stylus Profilometer, Bruker). The average duration of the print is 10 seconds per device thus enabling rapid manufacturing. The substrate to nozzle distance is fixed at 0.25 mm and the print is carried out at a pneumatic nozzle pressure of 0.15 Pa. A higher nozzle pressure would result in further reduction in viscosity due to the increased shear rate thus degrading the printing quality and uniformity. The nozzle speed is kept constant at 8 mm/s. On print

completion the printer bed is heated to 80  $^{\circ}\text{C}$  for one hour for solvent evaporation.

### C. Realising electrodes for DIW sensors

The response of tunnelling-based sensors is governed by the material, area and conductivity of the contact, as the charge carriers must be effectively captured by the collecting electrode. Additionally, the contact should be sufficiently adhered to the sensor without risk of damage or cracking during cyclic application of dynamic pressure. Herein, we have utilized the perpendicular tunnelling pathway by placing the electrodes on the side of the sensor with an additional controlled overlapping area near the top (Fig. 2). This provides for comparatively uniform capture of electrons. Highly conductive and low viscosity Silver paint (RS Pro Conductive Paint) is gently brushed against the dried sensor with the nozzle onto which copper wires are attached. The contacts are then annealed at 150 $^{\circ}\text{C}$  for 15 minutes to improve mechanical and electrical properties after which they are attached firmly to the substrate using polyimide tape. The fabricated device is shown in Fig. 2. In order to better analyse the morphology of the sensor, cross-section scanning electron microscopy (SEM) is carried out using the SU8240 FE-SEM (Hitachi Hitech) at an accelerating voltage of 5 kV. The layered nature of the graphite powder after printing can be clearly observed in Fig. 3.

## III. RESULTS AND DISCUSSION

The base conductivity of the sensors is measured using the four-point probe method (Ossila Ltd.). Three sensors have been fabricated and they are referred as S1 ( $R_0 = 170 \Omega$ ), S2 ( $R_0 = 182 \Omega$ ) and S3 ( $R_0 = 146 \Omega$ ). The average conductivity of the sensors is 92.66 S/m with a standard deviation of 2.05 S/m, leading to a coefficient of variation of only 2.21%. Furthermore, the as-fabricated sensors were subjected to cyclic loading and unloading during which their piezoresistive response and uniformity was determined. The piezoresistive response is given by  $\Delta R/R_0$  % where  $\Delta R$  denotes the difference between the measured two-terminal resistance and the initial resistance prior

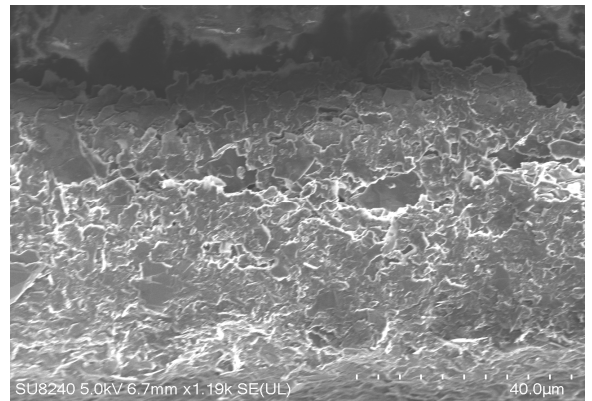


Fig. 3. Cross-sectional SEM of layered graphite-based sensor.

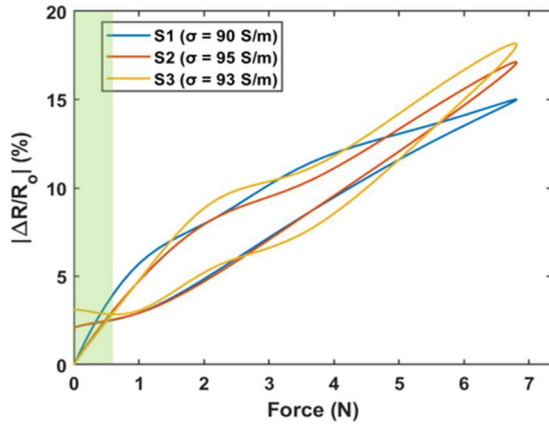


Fig. 4. Raw piezoresistive hysteresis curve of three sensors during comparably stable region of operation obtained directly from load cell. A force of 7 N corresponds to  $\sim 3.2$  MPa.

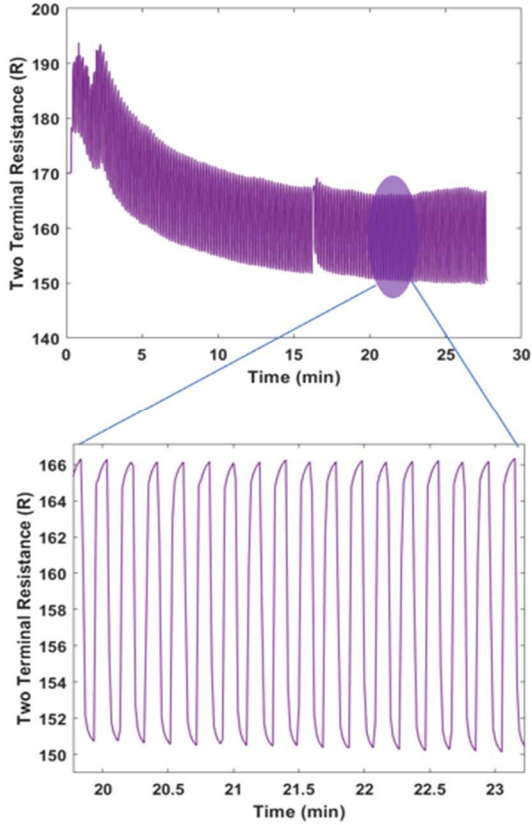


Fig. 5. Cyclic loading-unloading of piezoresistive sensor (resistance in Ohms).

to loading. The sensor is firmly attached to a single point load cell (model 1042, VPG Transducers) while a contact force of surface area  $19.6 \text{ mm}^2$  is periodically applied to the sensor via a high precision linear motor controlled by a square-wave frequency of 0.5 Hz. The low signal of the wave corresponds to 0 N and the high signal corresponds to approximately 7 N ( $\sim 3.2$  MPa). A custom-made LabVIEW (Keysight technologies) program is developed to record the output of the load cell and the piezoresistive sensor response synchronously using two digital multimeters (models 34461A and 34465A) for  $\sim 30$  minutes.

As the graphite composite is loaded, the conducting particles form tunnelling pathways reducing the measured resistance. Due

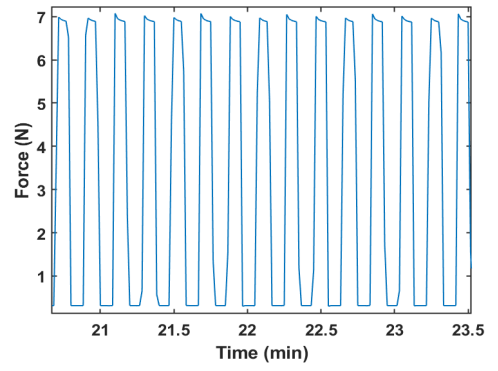


Fig. 6. Intrinsic hysteresis in response of load cell particularly during loading.

to the non-elastic nature of the sensor, hysteresis can be observed in the response of the sensor as shown in Fig. 4. Furthermore, the piezoresistive hysteresis of the sensors is shown in Fig. 4 wherein the green area of the curve represents the cross-over points of the loading and unloading cycles for each sensor. This hysteresis can also be seen in the extended view in Fig. 5 in which the resistance of the sensor exhibits finite rising and falling edges. Currently the sensor exhibits pseudo-plastic behaviour wherein the base-resistance of the sensor is not recovered after pressure is removed. This will be improved by intermixing the percolation with printing appropriate elastomers. This is in part due to the inherent hysteresis of the load cell during cyclic operations as shown in Fig. 6. The rise and decay time of the sensors is  $\sim 0.5$  s which can be considerably improved via electrode optimization to efficiently capture all tunnelling carriers. The average piezoresistive response is 16.76% at a force of approximately 7 N and a standard deviation of 1.19% leading to a coefficient of variation of only 7.1%. An interesting perspective provided by the sensors is that the piezoresistive hysteresis also occurs at lower forces, which correspond to grasping pressures. This is indicative of the sensor's response towards memory of the applied force and could potentially be utilised in the development of mechanical sensors with memory. Further analysis at higher frequencies and bending configurations will be carried out as the extension to this paper.

#### IV. CONCLUSION

In summary, we have shown the direct ink-writing based piezoresistive sensors and characterized their responses. The uniformity of the devices is gauged via the base conductivity and piezoresistive response, both of which exhibit a very good coefficient of variation of 2.21% and 7.1%, respectively. It is noted that the sensors are sensitive to a wide range of pressures from 0-7 N ( $\sim 3.2$  MPa). Further studies will be carried out to encapsulate the sensors for improved performance. The sensors will then be utilized in conjunction with transistors and actuators towards tactile based environment exploration, e.g., object grasping and manipulation.

#### ACKNOWLEDGMENT

M. S. B thanks Mr. Luca De Pamphilis for assistance during SEM imaging.

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