

On 3D printing of thermoplastic polyurethane over woven fabric for wearable sensors

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Abstract: In the past two decades several studies have been reported on thermoplastic-based 3D printed sensors. But little has been reported on wearable sensors prepared by thermoplastic printing on woven fabric, especially for four-dimensional (4D) properties. This study reports the 3D printing of thermoplastic polyurethane (TPU) on woven fabric by using a fused filament fabrication (FFF) setup for the fabrication of wearable sensors. Further, rheological, mechanical, voltage resistance (V-R), radio frequency (RF), and 4D capabilities of the 3D printed substrate have been established. The 3D printing of TPU on woven fabric was performed at 225°C nozzle temperature with a printing speed of 21 mm/s and infill density was kept at 80% and 100%. The RF characteristics were calculated using a vector network analyzer (VNA) which suggests that there is no shift in resonating frequency (2.60 GHz at -81dB) for ring resonator printed with 100% infill density (with an increase in strain value), whereas for 80% infill density sensor there was a notable shift in resonating frequency (from 2.82GHz at -75 dB to 2.71 GHz at -76 dB) when the sensor was strained under load which further lead to a change in dielectric constant (ϵ_r) and loss tangent ($\tan \delta$).

Keywords: Fused filament fabrication; sensor; mechanical strength; radio-frequency; 4D

Introduction: In the past two decades, a lot of studies have been reported on 3D printed sensors [1-3] with tune-able RF characteristics as 4D properties. The static behavior of 3D printing was made dynamic by adding time as the fourth dimension (by giving some stimulus resulting in a change in the behavior of the printed part) [4]. Shape memory polymers have shown great applications in 4D printing due to their ability to adopt the same shape again and again [5]. TPU is one of the most popular elastomers having good shape memory properties, biocompatibility, high elasticity [6], and good printability which makes it a suitable choice for 4D printing. Researchers have reported direct printing of polymers on fabrics resulting in hard surfaces providing free movement of fabric [7]. These printed parts have been used as substrates for 3D-printed wearable patch antennas by researchers.

Microstrip patch antenna (MPA) (as a sensor) is comprised of a substrate (dielectric material) having a conductive radiating patch and a ground part on the top and bottom side of the substrate respectively [7]. The MPA can be easily 3D printed and their miniature design and light weight have made them one of the most suitable materials for sensing applications [8,9]. The MPA printed on woven fabric are termed smart wearable textiles achieved by a combination of flexible MPA with traditional textiles, that may be further used for communication, sensing, and artificial intelligence [10]. Significant studies have been reported on the direct printing of polymer over fabrics [5-10]. Researchers have printed polymer on textiles to achieve a 3D effect using FFF [11]. It has been observed that while FFF, the polymer has shown good printability and mechanical strength on woven clothes as compared to knitted clothes [7]. The researchers found that for better adhesion the polymer should flow in the small gaps provided in the fabrics which can be attained from the polymers having high viscosity and pressure (which may be applied by providing less gap between the nozzle and bed) [12]. The technical challenges of inkjet-printed dipole antennas on textiles for wearable communications have been outlined by some researchers [12,13]. Along the same line, a 3D-printed hybrid wearable wrist orthosis has also been designed [14]. The majority of current pressure sensors are based on negative resistance fluctuations [15]. It is challenging to strike a compromise between sensitivity and detection range. The researchers have developed a flexible, low-cost pressure sensor based on laser-scribed graphene that exhibits a positive resistance-pressure response. For materials based on carbon fiber cement, the efficiency of resistance-based sensing has been demonstrated under compression, tension, and flexure. The volume resistance is calculated when subjected to uniaxial tension or compression. It has been reported that compression causes a decrease in resistance whereas tension causes an increase, and when the load is released, the effect is almost entirely reversible [16-18].

The literature review reveals that in the past two decades, several studies have been reported on thermoplastic-based 3D-printed sensors. But hitherto little has been reported on wearable sensors prepared by thermoplastic printing on woven fabric, especially for 4D characteristics (like tune-able RF properties). This study reports 3D printing of TPU substrate on woven fabric (lycra on the weft side and cotton on the warp side) by using an FFF setup (open source without any change in hardware/ software) for the fabrication of wearable sensors.

Experimentation: The methodology used in this research is shown in Fig. 1.

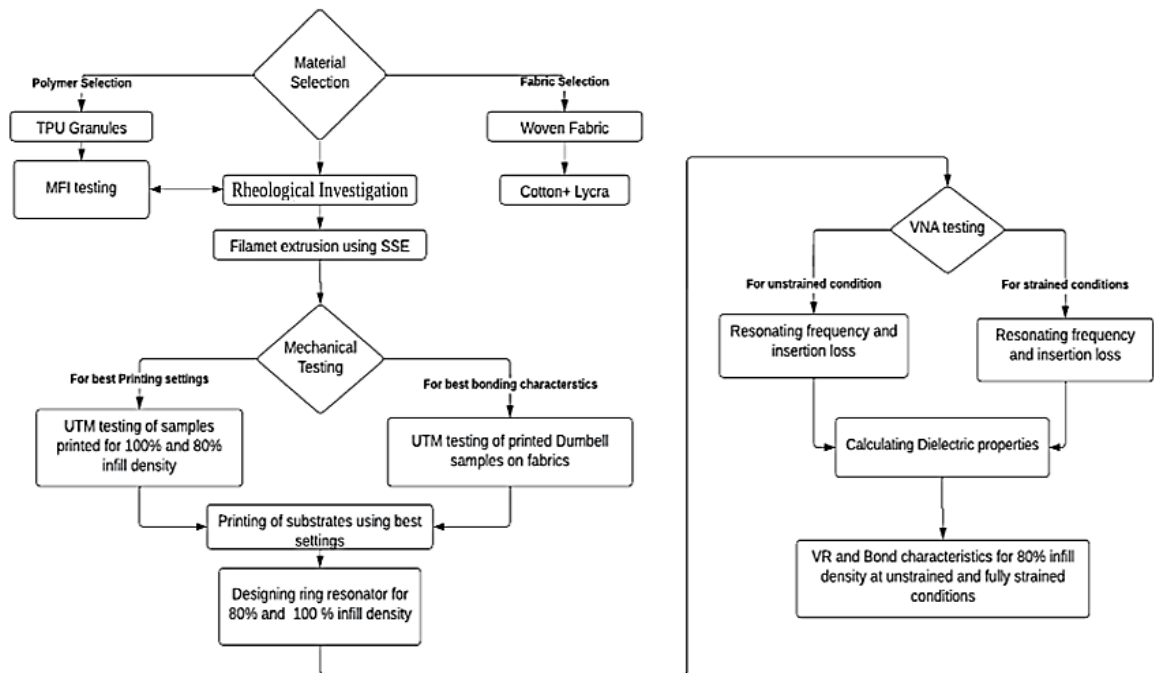


Fig. 1: Adopted methodology

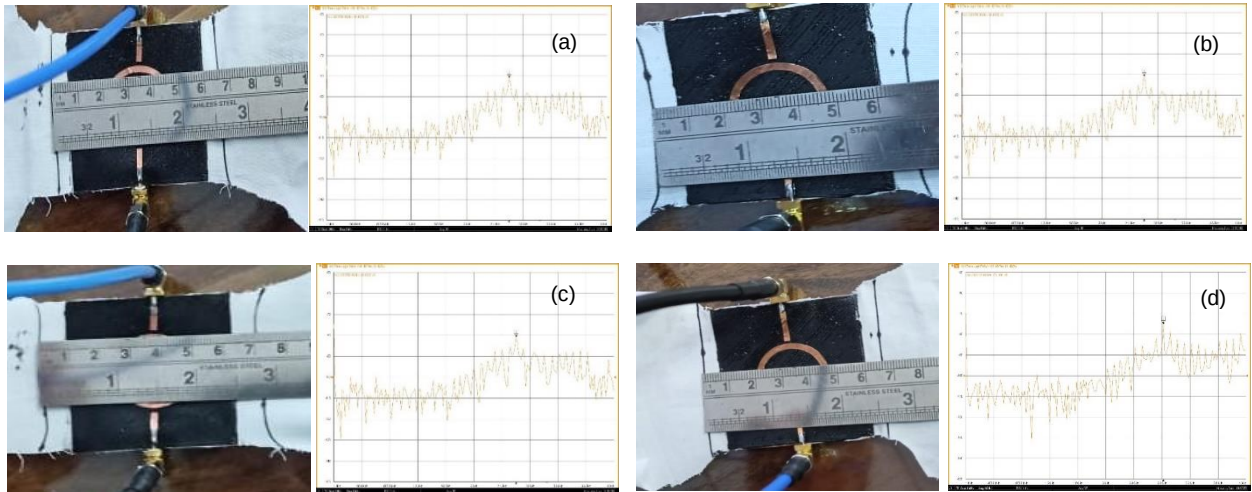
Literature shows the better printability of polymer on woven fabric as compared to knitted fabric [7]. In this study, the woven fabric with lycra fibers on its weft side and cotton fibers on its warp side has been judiciously selected so that the lycra provides the required stretchability in one direction of the fabric. TPU granules with Shore A hardness of 85A (as available in the commercial market) were selected due to low retraction, good printability, and high flexibility. The MFI for a selected grade of TPU was 29.7 ± 1.2 g/10min as per ASTM D 1238 (temperature 190°C under 2.16 kg load). Further, the filament ($\phi 1.75 \pm 0.05$ mm) was extruded using a single screw extruder (SSE) at 170°C screw temperature at 7 rpm. **The recycled TPU pallets were used in this study for recycling waste which may be extruded at optimized settings using a single screw extruder. This is in line with the concept of circular economy.** The printing characteristics of TPU filament were ascertained by printing it on FFF (Make: Ender 3 pro, open-source printer) and the selected settings were 225°C nozzle temperature, and 50°C bed temperature at a printing speed of 21 mm/s, at an infill density of 80% and 100%. Mechanical testing of the printed sample was performed and it was observed that the sample with an infill of 80% has shown less stiffness than the sample printed at 100% infill. Further, the TPU was printed directly on the woven fabric at the same selected settings of FFF. The ring resonator was designed using the equations used in previous studies [8]. After this, the substrate was printed on the woven fabric of size 54×60×0.65 mm. **The ring designed as per the previous study [8] was cut from Cu tape and pasted on the printed substrate as shown in Fig. 2.** Further for checking the RF characteristics, insertion loss (S_{21}) was checked on VNA at unstrained and fully strained conditions from which the dielectric properties (ϵ_r and $\tan \delta$) was calculated in unstrained and fully strained conditions. Further, these results are supported by attenuated total reflection (ATR)-Fourier transformed infrared (FTIR) and VR characterizations under unstrained and strained conditions.



Fig. 2: Front and bottom side of a ring resonator

Result and discussion

The tensile testing on a universal testing machine (UTM) outlined that with 80% infill density, a sample has low stiffness (3.504 N/mm) as compared to 100% infill density (4.05 N/mm). The prepared ring resonator for 100% infill density, when tested on VNA resonated at 2.60 GHz with an S_{21} of -81 dB. This sample has not shown any shift in resonating frequency under the strained conditions as shown in Fig. 3 (a), (b), and (c) whereas the ring resonator with an infill density of 80% has shown a prominent shift of resonating frequency under the fully strained condition. The resonating frequency was observed 2.82GHz with an S_{21} of -75 dB under unstrained conditions and 2.71 GHz with an S_{21} of -76 dB under the fully strained condition as shown in Fig. 3 (d) and (e). It was also observed that there was no frequency shift at the intermediate strain provided until it reached the fully strained condition as shown in Fig. 2(f). As observed in Fig. 3, for unstrained, partially, and fully strained conditions the length was 65, 69, and 72 mm respectively. It should be noted that for the fully strained condition, the strain was observed as 10.7%. The value of resonating frequency, ϵ_r , and $\tan \delta$ for each condition is shown in Table 1. **This work is a pilot study to ascertain whether the substrate with 100% infill density has 4D properties or not. It was ascertained that it does not show any 4D characteristics with selected stimuli (Fig.3). Then the infill density was reduced to 80% and it showed the 4D characterization on applying load. Further investigations may be performed with different infill density percentages.** To further understand this behavior ATR-FTIR analysis was performed by the incidence of IR rays ranging from 500-4000 cm^{-1} wavenumber (WN) on the surface of a 3D printed substrate of 80% infill density on stretched and unstretched woven fabric (Fig. 4 and 5).



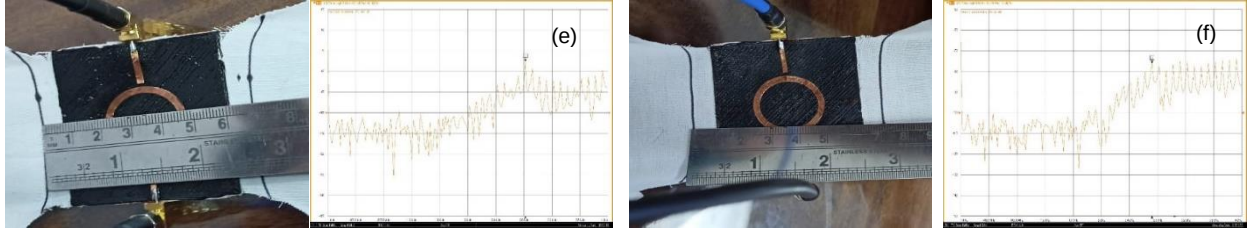


Fig. 3: RF characteristics for (a) 100% unstrained, (b) 100% partially strained (c) 100% fully strained, (d) 80% unstrained, (e) 80% partially strained, and (f) 80% fully strained

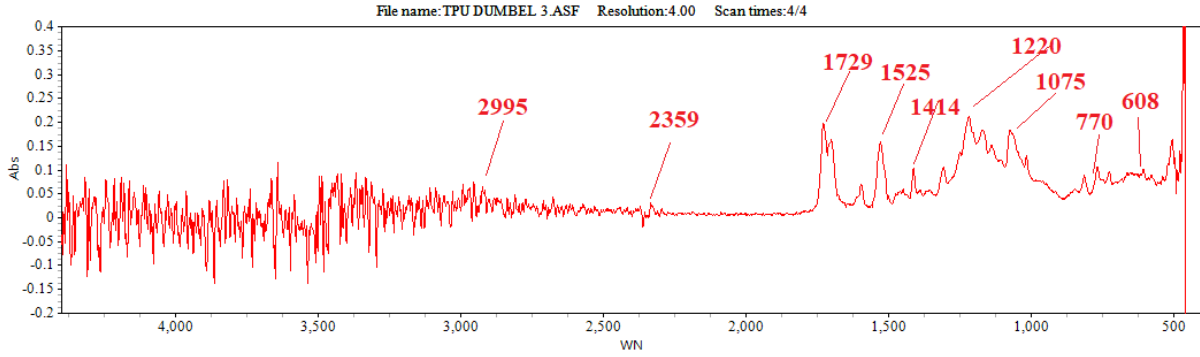


Fig. 4: FTIR for TPU printed on the unstrained woven fabric sample

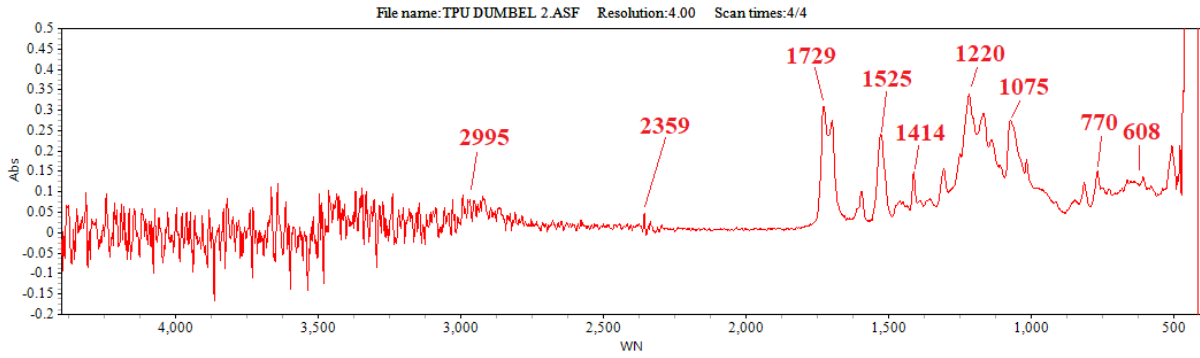


Fig. 5: FTIR for TPU printed on the fully strained woven fabric sample

Table 1: RF and dielectric characteristics of unstrained and fully strained samples

Samples	Resonating frequency (GHz)	ϵ_r	$\tan \delta$
Unstrained Sample 100% infill density	2.60	2.30	0.0044
Fully strained Sample with 100% infill density	2.60	2.30	0.0044
Unstrained Sample 80% infill density	2.82	1.87	0.0044
Fully strained Sample with 80% infill density	2.71	2.07	0.0044

The peaks observed in Fig. 4 confirm that the polymer is TPU [19-21]. A peak at 2995 cm^{-1} shows the asymmetric and symmetric stretching of -CH bonds, 2359 cm^{-1} defines the CO_2 stretched group, 1729 cm^{-1} , and 1525 cm^{-1}

indicate the urethane structure of -C=O bonding -N-H deformation respectively, 1300 to 1100 cm^{-1} spectral region is assigned in literature to C-O stretching vibrations of urethane units or C-O stretching vibrations of esters. It was observed that the absorbance of the sample increases with stretching (Fig. 5) which may lead to more charge storage resulting in a better ϵ_r . It may be noted that FTIR was performed to find the difference in the bond characteristics of the substrate under unstrained and strained conditions. The peaks in terms of the wave number are identical whereas there is a clear difference between the absorbance of the peaks. The absorbance of peaks in Fig. 5 is more than the absorbance in Fig. 4 of FTIR. Further, the VR characteristics were observed for stretched and unstretched samples. It was observed that the unstretched sample has shown 64.88Ω resistance at 1.5V. The same was reduced to 36.73Ω when the sample was stretched as shown in Fig.6 (validating the increment of ϵ_r with the decreased value of resistance [22]). In this study, a woven fabric (70% cotton (on the warp side) and -30% lycra (on the weft side)) was used for analysis. Further study may be performed for V-R curves of Lycra alone.

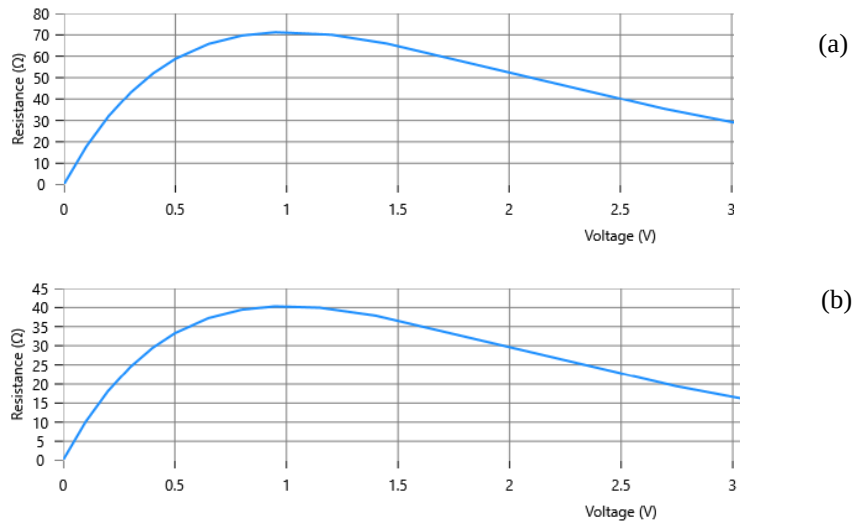


Fig. 6: VR characteristics of the unstrained sample (a), fully strained sample (b)

It may be noted that ϵ_r of a substance or material is a measure of its ability to store electrical energy. Mathematically it can be defined as the ratio of the capacitance of a capacitor with the dielectric material as the medium to its capacitance with a vacuum between its plates. The capacitance (C) is the ratio of the amount of charge (q) to the potential difference (V). So, it can be concluded that a higher charge will lead to higher capacitance which will further increase the ϵ_r . In this study, the aim is to ascertain how ϵ_r changes with the load applied to the substrate. To ascertain the safety of the human body the sensor-specific absorption rate (SAR) value has been simulated as per the reported study [23]. The SAR value was simulated using a high-frequency structure simulator (HFSS) (Fig. 7) as 0.004 W/kg over 1 g tissue which is well under 1.6 W/kg over 1 g tissue.

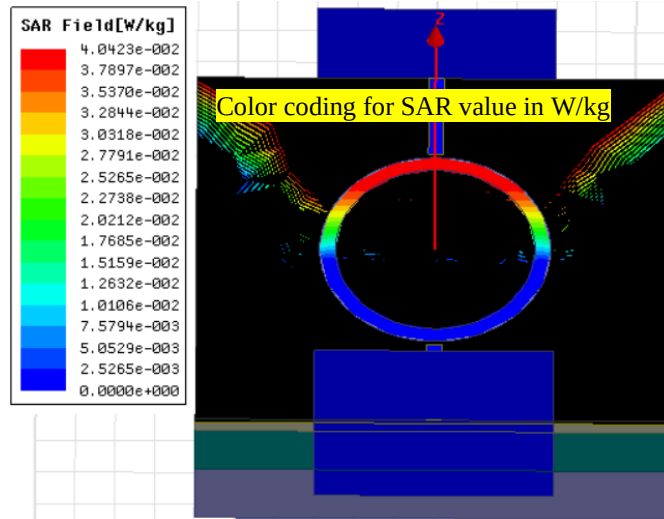


Fig.7: SAR value of the purposed sensor

For material properties of the TPU alone and TPU with fabric thermal characterization was performed using differential scanning calorimetry (DSC). Samples of TPU and TPU printed on CLW fabric were tested in a temperature range of 40-200°C with a rate of 10 K/min for 02 heating-cooling cycles as shown in Fig. 8. The first heating cycle showed that the glass transition (at first endothermic peak) of the TPU started at 40.01 °C to 42.25 °C whereas, for TPU printed on CLW fabric, the glass transition was observed from 49.01°C to 50.39 °C which shows a shift in the glass transition temperature denoting that the TPU printed on CLW fabric can be used at a little higher temperature in line with previously reported study [23] (Fig. 8).

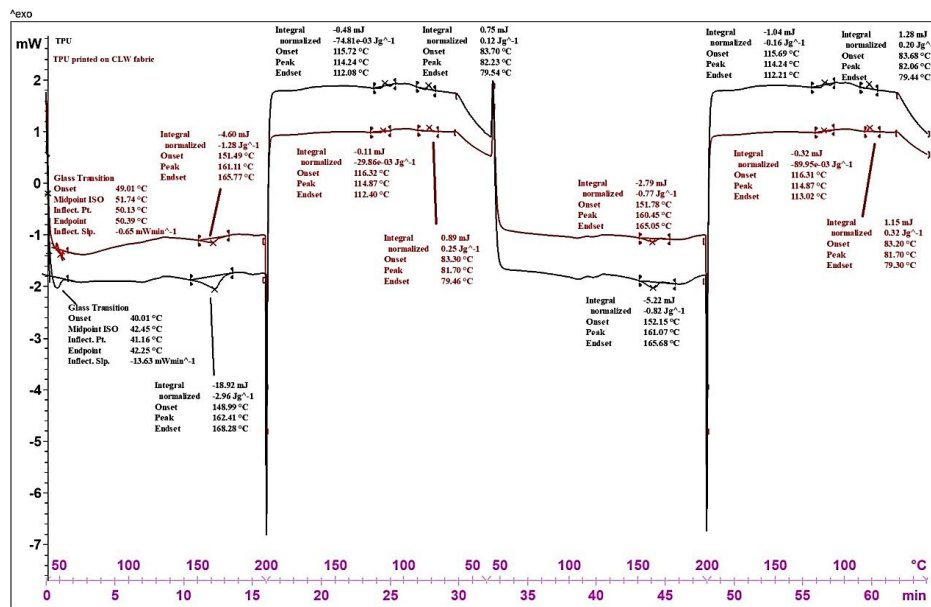


Fig. 8: DSC results for TPU and TPU printed on CLW fabric (Temperature ranging from 40°C to 200°C)

Mechanical testing was also performed for TPU alone and TPU printed on fabric (Table 2) and some improvement in Young's modulus (E) and stiffness was noticed, which may have significant advantages in sensing capabilities. The focus of this work was on the 4D characterization of the printed substrate using the ring resonator test. Further, the fabricated sensor may be used in Bluetooth or WiFi applications.

Table 2: Observed mechanical properties

Material	PS (MPa)	Stiffness (N/mm)	SE (N.mm)	E (MPa)
TPU alone	9.1±0.18	3.1±0.11	525.6±1.7	12±0.8
TPU printed on fabric	5.4±0.67	3.8±0.29	500.1±1.3	14±1.0

Conclusions

- The results of mechanical testing outlined that a sample with 80% infill density was less stiff as compared to a 100% infill density substrate providing more strain. The shift in resonating frequency due to the effect of strain caused by mechanical force during RF analysis justifies the 4D characteristics of the 3D printed sensor (for 80% infill density).
- The RF characteristics for the 80% infill density substrate outlined that when the sensor was strained under load, lead to a change in ϵ_r and $\tan \delta$ which was calculated as 1.87 and 0.0044 for unstrained conditions and 2.07 and 0.0044 under fully strained conditions ($\approx 10.7\%$) and gives the same value again ($\epsilon_r = 1.87$ and $\tan \delta = 0.0048$) when unstrained.
- ATR-FTIR analysis suggested that the absorbance of the stretched sensor was increased leading to an increase in charge storage capacity and ϵ_r . Also, it has been ascertained that with an increase in strain (along the length) in the substrate there was a marginal change in the width of a sensor, however, this resulted in improved ϵ_r without affecting $\tan \delta$ for 4D sensor applications.

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