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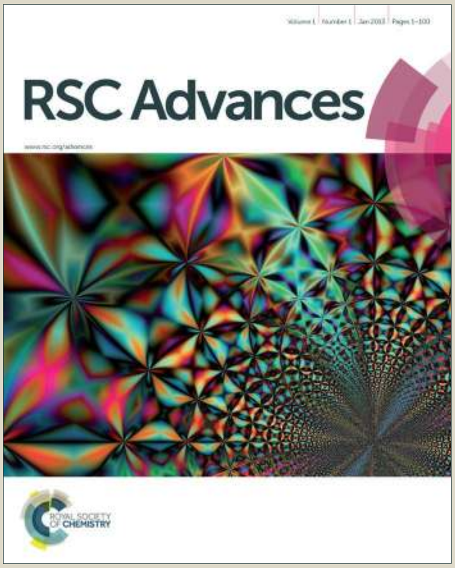

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Solvent-free fabrication of Multi-walled carbon nanotube based flexible pressure sensor for ultra-sensitive touch pad and electronic skin applications

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Abstract

This paper reports a solvent free, low cost fabrication and clean process of an ultrasensitive touch pad by sandwiching multi walled carbon nanotubes (MWCNTs) between bottom polyimide (PI) substrate and top cellulose paper using rolling pin and pre-compaction mechanical pressing technique. The sensing mechanism is due to pressing force dependent contact of MWCNTs between top cellulose paper and bottom PI. The recently developed solvent free pencil on paper approach for fabricating pressure sensors has the drawback of low throughput which hinders its applicability in commercial domain. Here in this work, entire fabrication process is scalable and could be integrated to large area for mapping spatial pressure distribution. The as fabricated sensor has sensitivity of 0.549KPa^{-1}, response time of <32 ms and low power consumption of <1.9 mW. Apart from measuring pressing, tensile and compressive forces, the sensor can identify acoustic vibrations from a loud speaker. The fabricated pressure sensor was further applied as artifical electronic skin with 3 x 4 pixel array wherein it was observed that measured spatial distribution was consistent with shape and location of the object. This proposed flexible touch pad fabricated by a low energy fabrication and clean proces

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technology paves way for future wearable electronics such as flexible touch pads and human-machine interfaces.

Keywords: solvent free fabrication; clean process technology; polyimide tape; cellulose paper; multi walled carbon nanotube pressure sensor; flexible touch pad

1. Introduction

Advancement in flexible electronics has enabled researchers to focus on wide range of applications such as supercapacitor, batteries, solar cells, transistors, and sensors[1-4]. Each of these developments focuses on a single technology that would enable new type of consumer products. Most of the consumer products would require a user interface or an input device to be integrated. These user interface devices when integrated onto human skin have major applications such as artificial skin for burn victims, electronic skin, robotics and human machine interface. In this regard, wearable and lightweight pressure sensors are gaining interest for their use as a user interface in future flexible foldable electronic devices with applications ranging from touch on flexible displays[5], touch pads[6], electronic skin[7] etc. Most of these applications require stable operation, high sensitivity, fast response time and low power consumption. Recently, various nanomaterials such as nanowires, nanofibers, nanorods, metal oxides, carbon nanomaterials[8-10] are being utilized for the design of novel and flexible pressure sensors. However, in general, the sensing element in these pressure sensors are integrated into the device using dip coating[11], spin coating[12], drop casting[13] wherein the nanomaterials are dispersed in various toxic solvents, surfactants for dispersion and stabilization. Moreover, prolonged drying times at elevated temperatures are required to remove solvents to obtain the desired electrical properties. Furthermore, poor solubility and possibility in the change of properties of the nanomaterials could affect the performance of the fabricated devices. The above mentioned solvent based approaches involve processing of un-ecofriendly

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chemicals which contaminate the environment and are hazardous to life. In this regard, solvent free approaches such as pencil on paper are gaining interest [14]. However, the drawback of the said approach is its very low throughput which hinders its applicability in commercial domain. It should be noted even though photolithography is neccessary to fabricate devices with smaller feature size, it is unsuitable for applications where factors such as cost, environment, energy consumption are concerns. Also, photolithography plays an important role in acheiving ultra high sensitivity for practical use, this work targets to fabricate solvent free devices with sensitivity comparable to those fabricated using lithography process.

In this work, we propose solvent free fabrication of pressure sensor using MWCNT as active element by sandwich assembly of MWCNT between bottom polyimide (PI) substrate and top cellulose paper using rolling pin and pre-compaction mechanical pressing technique. Rolling pin ensures the uniform distribution of MWCNTs over the exposed area and pre-compaction mechanical pressing ensures the deposition of MWCNT on polyimide. Shadow mask used for fabrication was prepared by the use of butter paper. Polyimide was chosen to be the substrate material as it has excellent tear resistance. Flexible pressure sensor demands excellent tear resistance wherein the substrates are pressed thousands of time in bended and folded states.Unlike conventional methods which use either bulk rigid metal or soft PDMS to cover the sensing material in pressure sensors, we chose cellulose paper as it is biodegradable, soft, flexible and has nanofiber like structure which allows for more MWCNTs to be in contact with the metal electrodes when pressed, thereby increasing the sensitivity of the pressure sensor. Finally the device was laminated with another PI tape to limit the interference of changes in temperature, humidity etc. in the device performance as MWNCTs are known to respond to such changes [15]. It should be noted that even though we used cellulose paper on top of MWCNTs film, PI tape was used for lamination because cellulose paper has low tear resistance which is not suitable for pressure sensing applications. Moreover, polyimide tape adhesive was used for cellulose paper to adhere to the MWCNTs. So polyimide tape served two purposes:

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One passivating the whole devices and other adhering of cellulose paper to MWCNTs. Polyimide insulation provides high tear resistance wherein keys can be pressed for thousands times without affecting the performance of the device. Depositing metals on patterned polydimethylsiloxane (PDMS) which serves as top or bottom electrodes in most pressure sensors require sophisticated cleanroom techniques such as sputtering and evaporation systems which are expensive and use toxic gases. Sputtering and evaporation systems require elevated temperatures, plasma to be generated which consumes lot of energy and time. Moreover the deposited metals are not mechanically flexible and thus restrict the performance of the device in folded and bended states. PDMS was not used in this work to ensure solvent free fabrication. Here in this method, the deposition is carried out by rolling pin and pre-compaction mechanical pressing which does not involve the use of elevated temperature or plasma generation making the fabrication process simple, cost effective and energy efficient. To date, pressure sensors are based on force induced changes in capacitance[16], triboelectricity[17] and piezoelectricity[18]. Majority of the pressure sensor reported are either capacitive or piezoelectric based with very few reports on flexible resistive based pressure/touch sensor[19-20]. The resistive based pressure/touch sensor offers advantages such as low energy consumption, relatively simple fabrication etc. Herein we report solvent free MWCNTs based flexible resistive pressure sensor with high sensitivity, fast response time and low power consumption.The fabricated flexible pressure sensor array was employed for ultra-sensitive touch pad and electronic skin applications wherein the array functions as a human skin having the ability to sense the touch feeling of human skin.

2. Experimental Section

2.1. Fabrication of the device

The MWCNTs patterns were obtained using solvent free method wherein, polyimide (PI) was used as a supporting substrate. PI tape with the adhesive side was used as an active area for fabrication. PI tape was cleaned with acetone and isopropyl alcohol (IPA). Stencil mask was

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prepared using butter paper with array of 3 x 4 keys, wherein each key had an area of 0.5 cm^2. The mask was aligned on top of PI tape. MWCNT was deposited on the PI substrate by roll pin and pre-compaction mechanical pressing and butter paper mask was peeled off from the PI tape's adhesive, thereby forming the MWCNT patterns of the touch pad. Cleaning PI tape with acetone and IPA reduces the adhesive strength thereby making it easier to peel off the butter paper mask.To ensure uniformity, rolling was performed in one direction and for each key, 200 rolling cycles and 2 mg of MWCNT were optimized for uniform film deposition. Variation of resistance of the rolled pin MWCNTs was relevant to the rolling pin cycles and weight of the MWCNTs. Different weight of MWCNTs were deposited on each key and it was observed that as weight of MWCNTs is increased there is non-uniformity in the film. Cellulose filter paper was placed on top of the MWCNT patterns by the use of another PI tape which also served for laminating the device. Complete fabrication process flow is as shown in Figure 1. Details regarding the materials and characterization techniques can be found in supplementary information (SI).

3. Results and discussions

Solvent free processes for fabricating sensors and electronic devices are gaining interest due to simplicity of device fabrication, ease of processing of sensing elements without the use of complex manufacturing processes and high end equipment which often require a lot of energy. Polyimide is a highly tear resistance polymer which makes it well suited for use in pressure/touch sensor applications. Here MWCNTs were sandwich between bottom PI substrate and top cellulose paper by pre compaction rolling pin and mechanical pressing. Optimization in terms of rolling pin cycles and weight of MWCNTs were done to ensure uniform MWCNT film. Variation of thickness of MWCNT film with rolling pin cycles is as shown in figure 2. The rolling pin cycles were varied from 50 to 300 cycles. As rolling pin cycles increases decrease in the MWCNTs film thickness was observed. This is due to continuous pressure applied which

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tend to stretch MWCNTs film over the surface of PI. It should be noted that pressure applied while rolling pin would also play an important role in determining the quality of the film, but in this case, rolling pin was perfomed by human hand wherein the pressure variations are difficult to control. To ensure uniform pressure, post rolling pin, pre-compaction mechanical press with pressure of 5Kg/cm^2 for 15 seconds was performed. Pre-compaction mechanical press decreases uniformity in the film caused due to pressure variations during rolling pin. Figure 2 shows variation of the resistance of MWCNTs with rolling pin cycles. For resistance measurement silver paste was used as contacts. As the rolling pin cycles increased an increase in the resistance of MWCNTs film and decrease in the thickness was observed. This could be due to three reasons: 1) As the rolling pin cycle increases MWCNTs film tend to stretch which decreases the thickness of the film which increase the length to thickness ratio. The process is a two-step process namely spreading and rolling. In spreading MWCNTs are spread on application of less pressure with the help of roll pin. In rolling, the pressure and frequency of the roll pin is increased. As the rolling pin cycles increases the pressure on the MWCNTs film increases thereby further decreasing the thickness. 28 % decrease in the thickness is observed when the rolling pin cycles are increased from 50 to 300. Increasing the rolling pin cycles above 300 deforms the substrate. 2) As adhesive is present on bottom of MWCNTs film, on increasing rolling pin cycles MWCNTs may penetrate more in adhesive which further decreases the thickness. 3) Penetration of MWCNTs in adhesive increases the insulating nature of MWCNTs thereby increasing the resistance. The increase in resistance may also be attributed to the defects induced in MWCNTs due to the pressure applied. To study the effects of pressure on MWCNTs Raman spectroscopy was performed as shown in fig 2. Typical raman peaks of MWCNTs were observed: D band at 1342 cm^{-1}, G band at 1576 cm^{-1} and G' band at 2708 cm^{-1}. It was found that on pre-compaction press MWCNTs induced some defects which can be verified by calculating the I_D/I_G ratio. For pristine MWCNTs I_D/I_G ratio was found to be 0.315 while for pre-compaction pressed MWCNTs I_D/I_G ratio was found to be 0.403. Each wall of MWCNTs are

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bonded to each other by weak van der Waals forces which merge together on application of pressure thereby increasing the sp^3 hybridized bonds in MWCNTs. It should be noted that even though pre-compaction press induces some defects in MWCNTs it does not totally modify the structure from sp^2 to sp^3 hybridization. Increasing the pressure of pre-compaction press not only stiffens the substrate but also induces more defects in MWCNTs film. Raman spectra for the pristine MWCNTs and pre-compaction pressed MWCNTs can be found in SI as fig S1. Different weights of MWCNTs (0.5mg, 1mg, 2mg and 3mg) were used for fabrication wherein it was observed that lower weights of MWCNTs (0.5mg and 1mg) were not sufficient to form a uniform film over an area of 0.5 cm^2. 2mg and 3mg of MWCNTs were sufficient for uniform film formation in the area of 0.5 cm^2. When large weights of MWCNTs were used, not all MWCNTs adhered to the substrate (polyimide adhesive) which results in the non-uniformity of the film. To avoid this, large rolling cycles were required to make the film uniform. In this work, 2mg of MWCNTs with 200 rolling cycles were optimized for fabrication of single key. Figure 3a) shows the FESEM image of MWCNTs in powder form and Figure 3b) shows mechanically pressed MWCNTs on adhesive side of PI tape without rolling pin. It was observed that MWCNTs becomes densely populated but does not form a film. Figure 3c) shows the morphology of 2 mg MWCNTs film after rolling pin for 200 cycles and then mechanical pressing with pressure of $5kg/cm^2$. Formation of film was observed which indicates that 200 rolling pins cycles are sufficient to form a film of MWCNTs on PI substrate. It was also observed that MWCNTs are well deposited on the PI substrate due to adhesive on top of PI tape. Figure 3d) shows the FESEM of 3mg MWCNTs thin film after rolling pin cycles for 200 cycles followed by mechanical pressing. No formation of film was observed when similar rolling pin cycles were performed for higher weights of MWCNTs. Thus, in case of higher weights of MWCNTs, more rolling pin cycles are required to make film uniform. Figure 3e) shows the FESEM image of cellulose paper wherein it is clearly observed that cellulose paper exhibits nanofiber like porous morphology. Figure 3f) shows the FESEM image of MWCNTs on

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cellulose paper. The FESEM image of MWCNTs on cellulose paper was obtained after successive hand press on the fabricated device and then peeling off the cellulose paper from the fabricated device. Due to the repeated hand press, deposition of MWCNTs on cellulose paper was observed. Therefore, whenever there is an external hand press, deposited MWCNTs on cellulose paper come in contact with MWCNTs deposited on PI substrate thereby increasing the density of MWCNTs connected between the metal contacts and hence there is increase in conductivity.

The concept of pressure/touch sensor developed was applied for fabricating flexible touch pad which was further integrated with LCD and LEDs to show its compatibility with advanced electronic devices. A button or key on touch pad is a physical device that exhibits binary states. When touched by a user it signals a binary change and returns to its original state when released. To form such an array of keys, we formed patterns on butter paper and transferred that patterns onto PI substrate using rolling pin and pre compaction mechanical pressing. Each key on the touch pad acts as a resistor whose resistance changes upon touch. The individual resistors have resistance that depends on the number of MWNCTs bridging between the metal contacts, which decreases, upon touch, as the number of MWCNTs bridging between the metal contacts increases. Increase in the number of MWCNTs bridging between the metal contacts increases the parallel connection of MWCNTs which contributes to the decrease in the resistance. Here, we consider human finger to give external pressure to the touch pad. Because of the lamination done by PI tape, human hand does not directly come in contact with the MWCNTs. When finger is bought closer to the touch pad and touched gently, it contributes to the increase in the number of MWCNTs between the metal contacts which makes detectable contribution to the decrease in the effective resistance measured across the key.

Keys/buttons fabricated using resistors require two contacts one of which is supplied to the voltage source and other to the ground. Herein, we fabricated mutually independent buttons wherein applied each key had two independent contacts for applied voltage and ground. The

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connections for applied voltage and ground were taken across MWCNTS by conductive copper tape, wherein all the applied voltage connections and all the grounds were given a single connection. The temporal response of 10 keys to repeated 30 presses of bare human hand can be found in SI as figure S2. It was found that all the keys had initial resistance in the range of 5-6.3KΩ and it decreased to 2.2 – 3.4KΩ range when pressed. Three such devices were fabricated and all the devices showed similar responses. This was due to precision in fabricating touch keys with precisely same amount of MWCNTs (2 mg/key), using the same mask in fabricating multiple touch pads. The sensing mechanism can be attributed to the force dependent contact of the MWNCTs between the top cellulose paper and bottom PI. MWCNTs are randomly arranged with not all MWCNTs connected to each other thereby forming potential barriers in between the MWCNTsdue to air or by the adhesive of the bottom polyimide tape. Whenever there is an external press the number of MWCNTs connected to each other increases, decreasing the potential barriers and thereby increasing the current. These potential barriers contribute to high resistance in relaxed state. As the pressure is applied, the number of MWCNTs connected to each other increases overcoming the potential barriers and creating more conduction paths. As soon as the pressure is released the MWCNTs come to their initial position thereby reforming potential barriers. Unlike bulk rigid metals, cellulose paper is soft flexible and has nanofiber like structure. The number of MWCNTs bridging depends on the external forces applied. On applying an external press, a small compressive deformation of cellulose paper enables more MWCNTs in contact with the copper wire thereby leading to more conductive paths. This causes an increase in current when a fixed voltage of 1V was applied. On removal of the external press, both cellulose paper and PI recovered back to their original shapes thereby reducing the amount of MWCNTs bridging the copper tape, thereby, leading to decrease of the current.

Figure 4a) shows the graph of initial resistance of the individual key and the resistance of the key when pressed by human hand. Measurements were taken for 3 devices and all the devices

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showed similar responses. Figure 4b) shows I-V characteristics of the key# 3 for different pressures and it was observed as applied pressure increases current increases. Applied pressure was kept constant while sweeping the voltage from -1V to 1V. Figure 4c) shows the temporal response of the touch pad with low frequency of human hand touches. It was observed that on a hand press there is a sudden rise in the current level and as soon as the hand is moved away the sensor does not returns to its initial resistance. This can be attributed to the fact that as the pressure is released there is not exact reformation of the potential barriers and hence current decreases to larger value than in unpressed/relaxed state. In addition to the pressing forces, the as fabricated sensor can simultaneously be used to detect bending forces. Under bend, which can also be treated as compression strain, the number of MWCNTs bridging would increase thereby increasing the conductivity.The response of the sensor under dynamic bending can be found in SI as Figure S3.

Sensitivity of the sensor was calculated as $S = (\Delta R/R_0)/\Delta P$, ΔR is relative change in resistance and R_0 is the resistance of the sensor under no load or press and ΔP is the change in the applied pressure. In this case, pressure was applied by air compressor whose pressure can be varied. The compressed air with a fixed pressure was pointed towards the keys of the touch pad and the corresponding change in the resistance was observed. As pressure of the compressed air was increased, decrease in the resistance was observed. Figure 4d) shows the graph between $\Delta R/R_0$vs ΔP, wherein the values were measured for all the 10 keys at different pressures and standard deviation with error was plotted. As shown in Figure 4d), a linear fit of graph between $\Delta R/R_0$vs ΔP, the sensitivity of the sensor was found to be 0.549KPa$^{-1.}$ According to our knowledge, the value is higher than the reported sensitivity values from other pressure sensors and second next to the recorded high sensitivity value in organic transistor pressure sensor wherein PDMS in conjuction with polymer was used for assembling separate layers via lamination[21-22]. It is worth noting that while complex fabrication steps and use of PDMS

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attributed to the high sensitivity of the pressure sensor in their case, the focus of our approach is on solvent free, energy efficient, low cost and simple fabrication while ensuring a very high sensitivity.

To investigate the reliability of the sensor the as fabricated sensor was applied with series of pressures from 1KPa to 5KPa. Figure 5a) shows the temporal resspnse of the pressure sensor under various pressures. Over the wide range of pressure applied current level increases with increase in the pressure. The response and relaxation curve of pressure sensor measured for ~10000 loading/unloading cycles of 2KPa are showin in figure 5b). During the large number of iterations performed, no difference in the current levels was observed demonstrating very high reproducibility of the pressure sensor. Fig 5c) shows the response of the pressure sensor for loading/unloading cycles at 2KPa for first 120 seconds of the measurement. The pressure was applied for 20 seconds and then a wait period was observed 15 seconds wherein no pressure was applied. The same was repeated for three times demonstrating excellent repeatability of the pressure sensor. It was then followed by loading/unloading cycles for ~10000 cycles. Fig 5d) shows the enlarged view of the response under loading/unloading cycles at 2KPa demonstrating very highly stable response. For response time, the sensor was subjected to very high frequecny touches of human hand and then difference in the time taken by the sensor to reach from 10% to 90% of maximum value was calculated. The as fabricated sensor was found to have response time of < 32ms.The graph showing the calculation of response time can be found in SI as fig S4.

To measure the change in resistance of the keys when pressed and to demonstrate interactive applications using the as fabricated touch pad we used Arduino platform. Arduino is an open source platform for signal processing and computation which has recently gained attention for its utility for applications in flexible and wearable electronics. To demonstrate bendable nature the as fabricated device was folded at one end and was tested for the pressure sensing at the other end. Figure 6a) shows the working of the flexible touch pad in folded state, wherein key

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#1 responds to the touch of the finger even when the device is in folded condition. The fold at the other end of the sensor did not have any effect on the performance of the device. Moreover, the whole device was folded and unfolded to test the reliability of the sensor. Interestingly the senor displayed similar response after unfolding. To further demonstrate the flexible nature the as fabricated sensor was mounted on curved surface of water bottle as shown in Figure 6b). As can be seen from fig 6b) the device responded in bend state when key# 1 and key#6 are pressed simultaneously thereby verifying the fact that the device can be integrated onto curved surfaces of human body. Each LED in the image corresponds to a key in the touch pad. Current limiting resistors of varying resistances were connected in series with the keys so that varied output for an individual key of the touch pad can be obtained. LEDs are connected to the digital pins of the Arduino whereas touch pad keys with resistance in series are connected to the analog pins of the Arduino. The resistance changes on the key press are converted to the voltage changes at the analog pins of the Arduino and are correspondingly coded for the digital pin to go high when the voltage across the corresponding key crosses threshold. Detailed schematic of the connection of the Arduino with the flexible touch pad and the underlying circuitry can be found in SI as fig S5. The images in Figure 6 depicts the touch pad capability of working in folded and bended state without having significant variation in the performance of the sensor.

To further demonstrate the functionality of the touch pad, we integrated the touch pad with the commercial available LCD. Video demonstrating 0 to 9 number press with the flexible touch pad can be found in supplementary information as Video S1. Further, we tested the integration of touch pad with commercial LCD for displaying the alphabets "IITH" which can be found in supplementary information as Video S2.

The sensor was further investigated for both tensile and compressive strain, wherein the sensor was bend inward and outward and the corresponding change in the resistance was measured. Since the polyimide tape has a poor stretchability, we explored the strain induced due to bending

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rather than stretching which has applications in human body movement monitoring. Under tensile stress (bending outward), the number of MWCNTs bridging between the metal contacts decreases thereby decreasing the conductivity. Under tensile strain, the MWCNTs tend to move away from each other and hence there is loss of conduction path which leads to decrease in the conductivity. On the other hand, under compressive stress, the MWCNTs tend to overlap with each other forming, thereby increasing the number of MWCNTs bridging between the metal contacts causing rise in the conductivity. Schematic demonstration of tensile and compressive strain and the temporal response under tensile and compressive strain respectively can be found in SI as fig S6. Pressure sensor along with the functionality of strain sensor is useful for electronic-skin application as strain sensor detects the human movement monitoring and pressure sensor senses the touch. The above results suggest that that the fabricated sensor can be further used for strain sensing applications such as healthcare, robotics, and human machine interface (HMI).

To further explore the potential applications of the pressure sensor, it was used to sense acoustic vibrations originating from sources such as loudspeaker. This application could further increase the functionality of the sensor to be used as an electronic-skin as it would be able to sense acoustic wave and also the air flow, which is of great interest to the burn and acid victims. To demonstrate such a capability, speaker was positioned close to the sensor with distance varying from few to tens of millimeters. Interestingly, pressure sensor responded to the acoustic vibrations forces from loudspeaker. The acoustic energy is the waves of varying sound pressure which causes the deformation in the cellulose paper and hence the number of MWCNTs bridging between the metal contacts increases, thereby increasing the conductivity. The schematic representation of the experimental set up for acoustic vibration detection and the response of the sensor to the acoustic vibrations from speaker can be found in SI as fig S7.

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Combining the multi-sensory applications of the fabricated sensor i.e pressure, strain and acoustic wave detection, the fabricated pressure sensor array was applied as artifical electronic skin with 3 x 4 pixel array. The pressure sensor array functions as a human skin having the ability to sense the touch feeling of human skin owing to its high sensitivity. Here each key acted as one pressure pixel to measure the pressure signal in form of resistance change. As shown in fig 7a), a pen was placed diagonally on the key numbers,“7“, “5“ and “3“ to test the sensor response to the external pressure. The output of the resistive variation in each pixel was then recorded and plotted via color contrast mapping as shown in fig 7b). It can be clearly seen that measured spatial distribution of the pressure is highly consistent with the shape and location of the pen, demonstrating that the touch key pad could be used as artifical electronic skin.

Till date, electronics for touch pad are either mechanically incompliant or are too expensive to be considered disposable. Touch pads that are commercially available are rigid in nature. The commercial keyboard or touch pad has two conducting layers which are separated by plastic sheet except at the holes where the keys push down to make the two sheet touch. It works on the digital logic of two binary values “0” and “1”, where “0” represents no press and hence no contact between the upper and lower conducting layer, and “1” represents press where there is contact between the sheets and current flows. It needs a physical key that connects the upper and lower conducting sheets when pressed. Moreover, it needs small rubber pieces such that it stops the key from moving down and pushes it back up when you release it acting like a spring. Furthermore, due to its rigid nature, it cannot be integrated to human skin for versatile applications. In our case, we fabricated a facile, solvent free pressure sensor that detects the press based on the resistance change and flexible nature of the materials used for fabrication. Due to the flexible nature of the device it can be integrated onto any substrate for diverse applications. Overall cost of the device (3 x 4) as estimated by the market prices of the components used is approximately US $ 0.51. It should be noted that the process mentioned can be optimized and automated wherein the rolling pin can be performed by programmable electric

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motor to get better repeatibility in the device performance. Also, the feature size is limited and depends on the precision with which the butter paper mask can be fabricated. Combining lithography to fabricate mask and automation in rolling pin can greatly reduce the feature size of the device as well as the sensitivity of the sensor.

There are several reports on flexible pressure sensors as touch pad and electronic-skin using various substrates and materials such as metallized cellulose paper[6], metal strain gauges on polyimide using inverted fabrication technique[23], MWCNTs with PDMS[24], Ag nanowires and PDMS with the use of tissue paper[8], CNTs on chewing gum[25]. However, the fabrication of those sensors makes use of processes such as photolithography, sputtering and evaporation which are not only expensive but consume a lot of energy. Moreover, the sensing element in these type of pressure sensors require methods like magnetic stirring, ultra-sonication for dispersion that make use of toxic solvents, thereby affecting the overall efficiency of the device. Recently, solvent free approaches on cellulose paper for fabricating sensors are gaining interest such as use of pencil on paper [26]. The major drawback of such fabrication process is its low throughput which hinders its applicability in commercial domain as each sensor has to be fabricated independently and then integrated to form an array. Another issue with using the paper substrate is its low tear resistance which makes it non-robust and decreases the life of the touch pad or pressure sensor. Cellulose paper for use as a disposable electronics is an ideal choice for the substrate material, but, in this work, we target applications which might use reusable electronics for interfacing the touch pad with personal computers, laptops, cell phones, gaming console etc. using standard protocols for communication. Other substrates and materials involve the use of energy inefficient, expensive microfabrication techniques which involve the use of toxic gases and chemicals.

4. Conclusion

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Here we report the use of ecofriendly and biodegradable cellulose paper and PI substrate with MWCNTs as sensing element for fabricating ultrasensitive pressure, strain and acoustic wave sensors. Combining the individual sensing platforms, array of sensors were fabricated without the use of clean room lithography technique for flexible touch pad and electronic skin applications. The deposition of MWCNTs on PI was performed using rolling pin and pre-compaction mechanical pressing which doesnt require elevated temperatures and plasma generation unlike in sputtering and evaporation technologies. Furthermore it doesn't involve any solvents or surfactants required in proceses such as drop casting, inkjet printing for fabricating nanomaterials based pressure sensors. The as fabricated sensor had sensitivity of 0.549KPa^{-1}, response time of <32 ms and low power consumption of <1.9 mW. The use of PI as a substrate is recommended for fabricating flexible touch pad because of its excellent tear resistance. The entire fabrication process is scalable and can be extended to other functional materials as well without the need of any complex and expensive equipments. The proposed flexible touch pad has enormous potential in future flexible electronics, biomedical devices, artifical electronic skin, smart packaging and gaming applications.

Supporting Information

Supporting information contains materials and characterization details, temporal response of as fabricated sensor for human hand press for 30 times for all 10 keys, temporal response of pressure sensor under dynamic bending, layout of the interface of fabricated touch pad with Arduino board, experimental set up for acoustic wave detection using the fabricated pressure sensor and videos demonstrating number (0 to 9) press and display of the alphabets "IITH" on commercial LCD.

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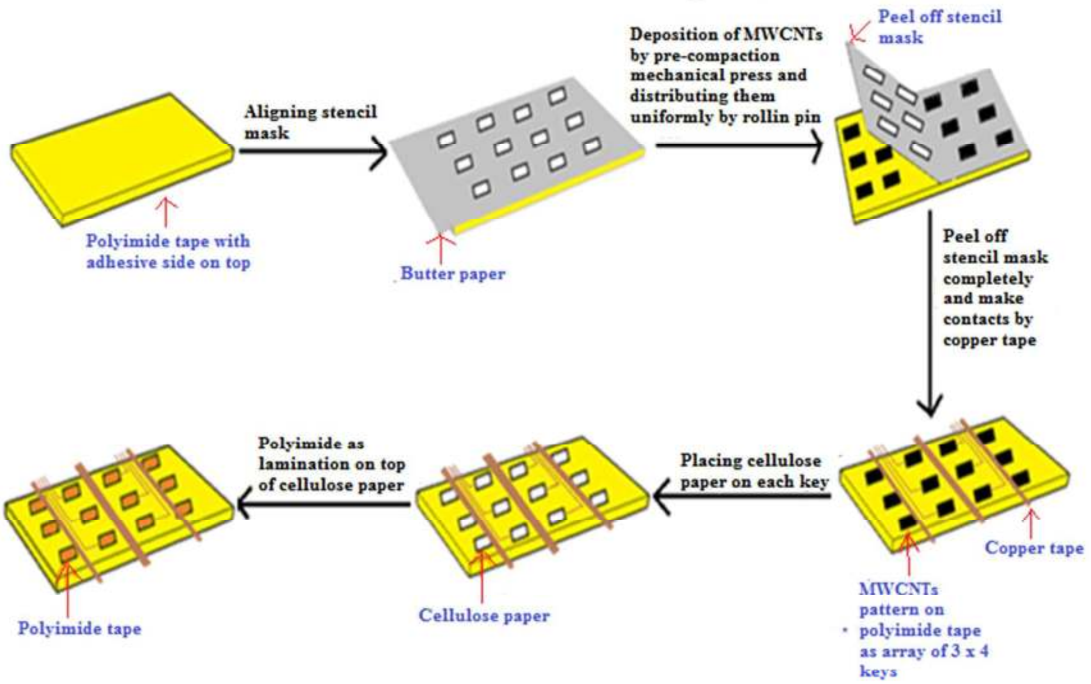

Figure 1.Schematic of solvent free fabrication procedure for MWCNTs based flexible touch pad

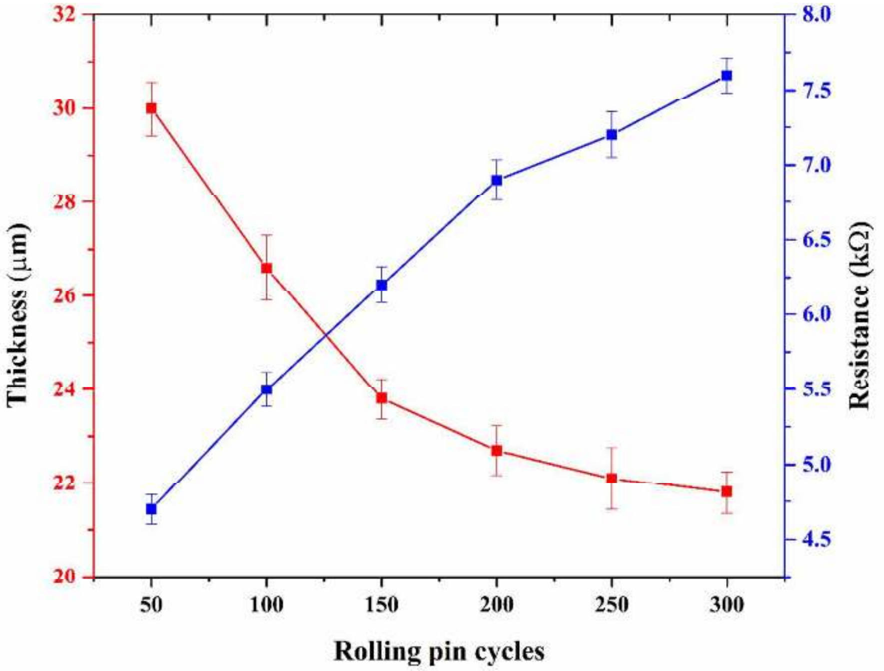

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Figure 2. Graph of rolling pin cyles v/s thickness of MWCNT and resistance of the MWCNT film for N =3 devices. As rolling pin cycles increases decrease in thickness and increase in resistance of MWCNT film was observed.

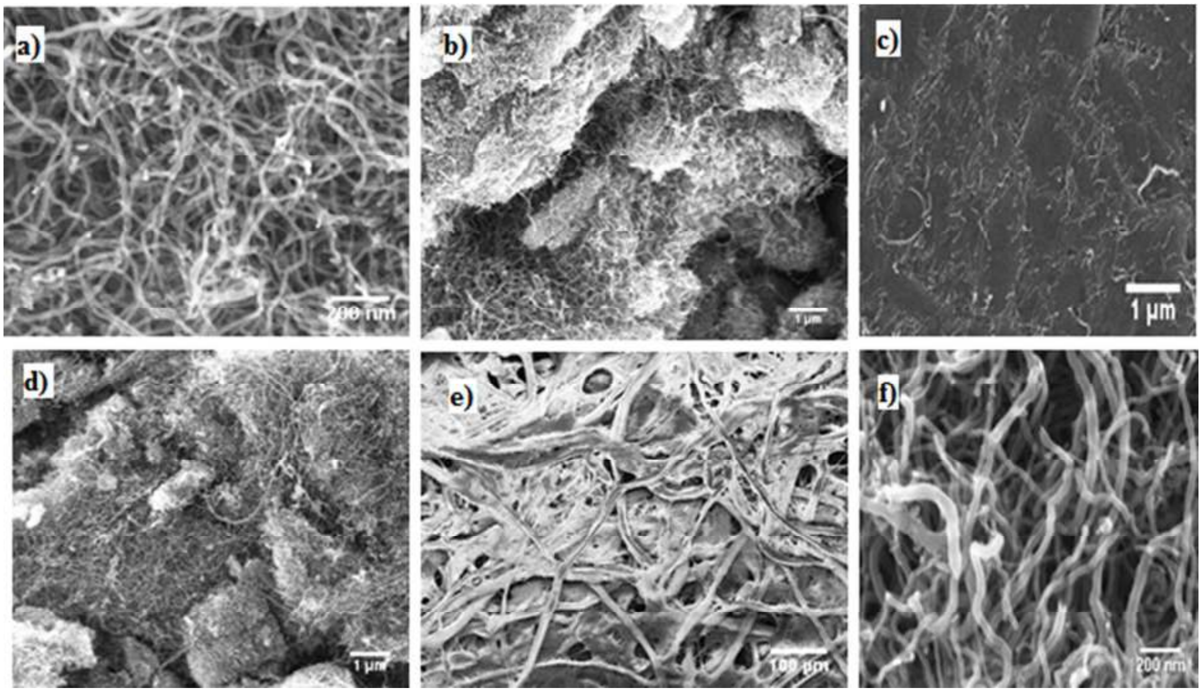

Figure 3.FESEM images of a) MWCNT powder b) mechanically pressed (without rolling pin) MWCNTs on PI substrate showing the densely populated MWCNTs c)mechanically pressed and 200 rolling cycles of rolling pin for 2mg of MWCNTs on PI substrate showing uniformity in the film d) mechanically pressed and 200 rolling cycles of rolling pin for 3mg of MWCNTs on PI substrate showing non-uniformity in the film e) cellulose paper showing the nanofiber like morphology f) MWCNTs on cellulose paper removed by peeling off cellulose paper after repeated touches on fabricated device.

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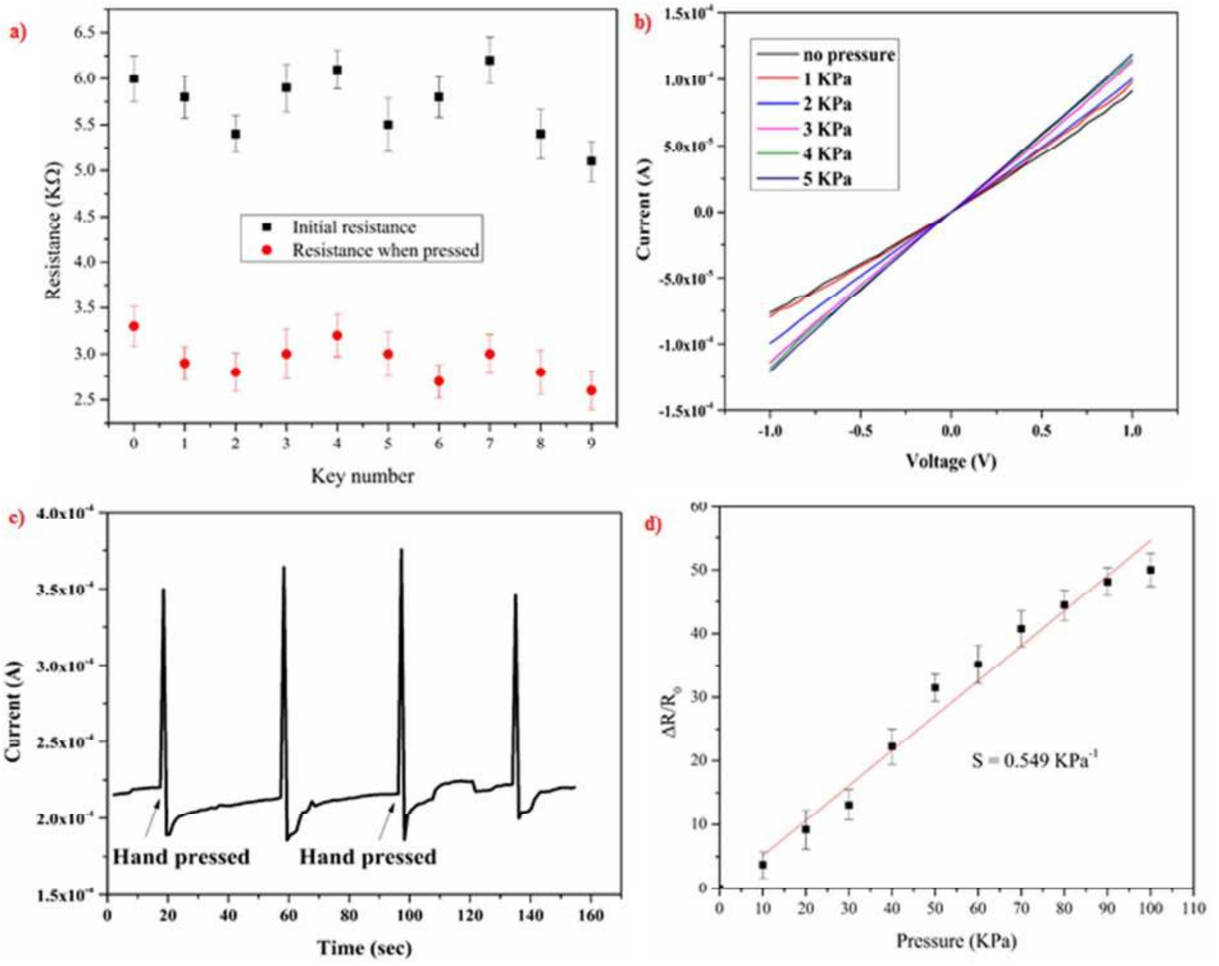

Figure 4. a) Graph showing the initial resistance (without human hand press) and resistance after human hand press for N=3 devices b) I-V characteristic of Key #3 at different pressures c) temporal response of low frequency touches of human d) Graph of $\Delta R/R_0$vs ΔP for calculating the sensitivity of the sensor. Values were measured for all 10 keys at different pressures. With linear fit of the graph sensitivity was calculated to be 0.549KPa^{-1}.

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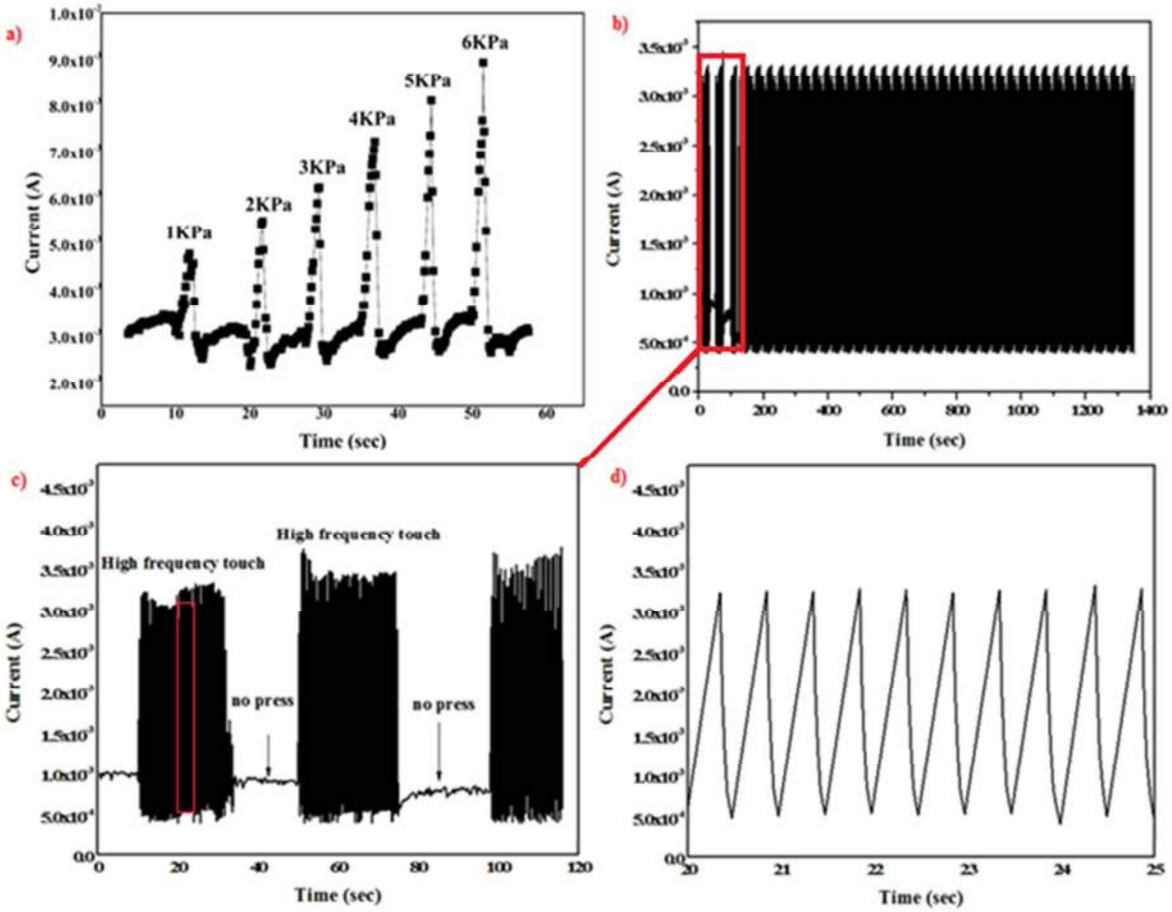

Figure 5: a) Graph showing the temporal response of the pressure sensor under various pressures ranging from 1KPa-5KPa b) Response of the pressure sensor for ~10000 loading/unloading cycles at 2KPa c) graph showing the response for first 120 seconds of the measurements where the loading/unloading of pressure was applied in iterations demonstrating excellent repeatability of the sensor d) Enlarged view of the response (red rectangle in fig 5c) showing highly stable response.

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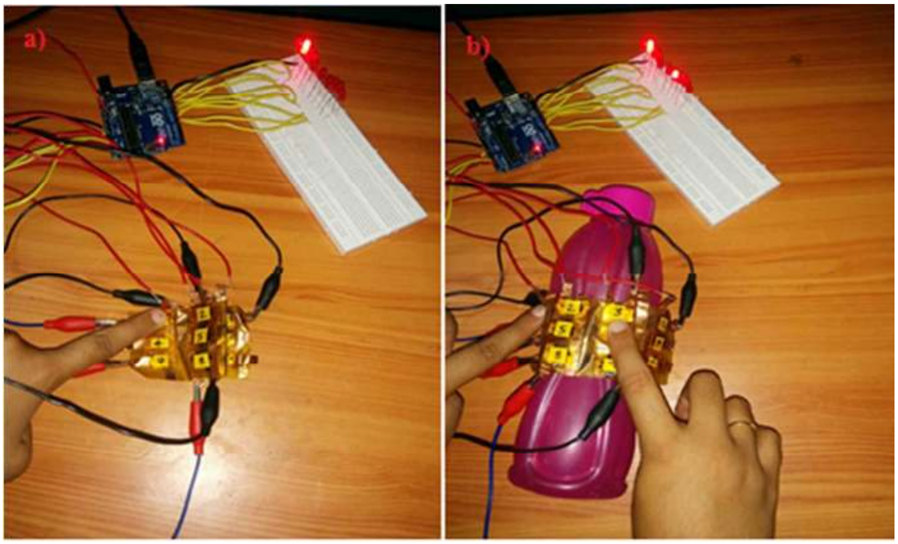

Figure 6. Photographs of a) working of flexible touch pad in folded state and its response to touch at key “1” b) flexible touch pad mounted on curved surface of water bottle and its response to touch at keys “1” and “6”.

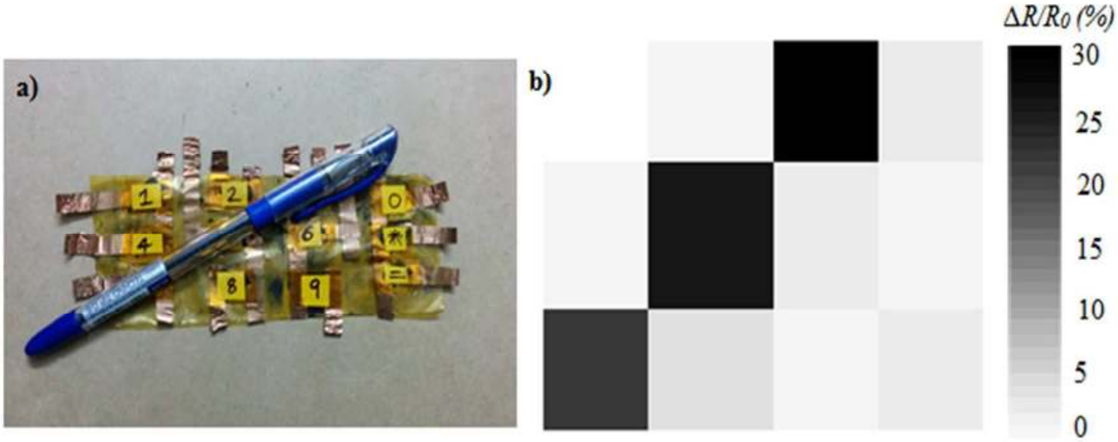

Figure 7. a) Photographs of the pressure sensor array of 3 x 4 with pen aligned diagonally on numbers “7“, “5“ and “3“ to test the pressure-sensing ability b) mapping profile of the pixel signals generated by pen.

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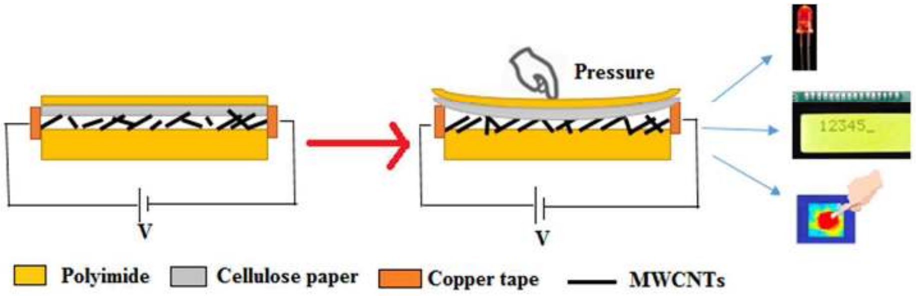
Graphical abstract



Schematic of the ultrasensitive touch pad by sandwiching multi-walled carbon nanotubes between polyimide and cellulose paper by rolling pin and pre-compaction mechanical pressing and its applicability as a user interface in modern electronic devices.