# Multifunctional Flexible PVDF-TrFE/$BaTiO_3$ Based Tactile Sensor for Touch and Temperature Monitoring

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***Abstract*— This paper presents an enhanced piezoelectricity based sensor for touch and temperature sensing. The sensor is realized over flexible polyimide film, making it suitable for application like e-skin. The sensing material is composed of Polyvinylidene Fluoride-Trifluoroethylene (PVDF-TrFE) and Barium Titanate ($BaTiO_3$) nanoparticles. While, the piezoelectric polymer PVDF-TrFE ensures the flexibility of sensor, BaTiO3 imparts high sensitivity to touch and temperature. The sensor is tested over temperature range which is common in daily life and the sensitivity to touch is characterized by tapping mode using fixed load. The results confirms the advantage of using poly-ceramic composite over piezoelectric polymer.**

***Keywords*— *flexible electronics; e-skin; tactile sensing***

## I. INTRODUCTION

Recently, extensive efforts have been taken to develop sensing systems for robots. In this area, one of the most important focus is the design and fabrication of electronic skin or E-skin for humanoid robots [1-3]. The successful implementation of E-skin relies on the improvement of its sensitivity and mechanical flexibility. The fundamental sensing capabilities of skin encompass the touch sensing, temperature sensing and so on [4]. So, for a quintessential E-skin, many tactile sensors and temperature sensors etc. are required to be realized on a support with enough flexibility to cover a conformable surface [5].

Piezoelectric materials are most common choice for the applications where dynamic sensing is required. In addition, due to present of temperature dependent polarisation, some piezoelectric material can be used for temperature sensing. In this regard, material such as $BaTiO_3$ and lead zirconium titanate (PZT) with high piezo and pyroelectric constants are used in a variety of applications. Especially, lead free $BaTiO_3$ based ferroelectrics are strong candidates for piezoelectric transducers due to their large permittivity, polarizations, and induced strains [6]. Moreover, $BaTiO_3$ film and particles has widely been used as humidity sensors and gas sensors because of its property to provide adsorbing site on grain surface to form active groups as surface charge carriers [7, 8].

However, ceramics are inherent to rigidity, fragility and poor processibility. On the contrary, ferroelectric polymers, such as poly (vinylidene fluoride) (PVDF) and its copolymer PVDF-TrFE owing to its flexibility, easy processibility and low mechanical impedance have been used as sensing material in variety of applications. For example, implantable pressure sensor for physiological signal monitoring and tactile sensor for prosthesis and robotics has been reported in literature [9-11]. Nevertheless, the low pyre and piezoelectric constants of PVDF-TrFE and the requirement of poling for polarizing thick film put hurdles in its effective utilization.

In this scenario, hetero-structural materials, such as polymer ceramic composites have received a significant attention because of their improved properties over the individual constituents. In addition, the properties of composite can be tailor made to suit the application by changing the proportions of the constituents. The ability to combine high piezo and pyro properties of ceramics with polymer's flexibility and easy processibility make poly-ceramic composite a good choice as transducer in flexible electronics application. Such composites based piezo materials could find application as electromechanical sensors, medical instrumentation, robotics, ultrasonic or underwater transducers, audio applications and so forth.

## II. FABRICATION

All solvents and chemicals were used as received. The $BaTiO_3$ nanoparticle with an average size of about 100 nm nanoparticle was purchased from Sigma Aldrich. The PVDF-TrFE copolymer pellets with a VDF/TrFE molar ratio of 70/30 was acquired from Piezotech (France).

PVDF-TrFE pellets (5 g) was dissolved in 50 mL of MEK solvent to make 10 wt% solution. The mixture was magnetically stirred at 70°C for 1 hour to get clear solution. 0.5 g of BT nanoparticles was separately dispersed in 10 mL of MEK using probe sonication for 20 mins. The two solutions are then mixed in 10% volume fractions and bath-sonicated for 1 hour to ensure uniform dispersion of nanoparticles in PVDF-TrFE. Polyimide film from Dupont was used as flexible substrate for the sensing device. The PI film is thoroughly cleaned via series of ultrasonic baths, dried and fixed on carrier wafer. 100 nm of Aluminum is evaporated using MEB550S as bottom electrode. The solution of PVDF and $BaTiO_3$ is then spin coated at 1000 rpm for 1 min. The spin-coated film was dried at 90ºC for 30 min to remove solvent. Then, film was annealed at 150ºC in nitrogen ambient for 3 hour to enhance film quality. The thickness of film was measured to be 4 µm after annealing. 10 nm/50 nm of nichrome and gold was then evaporated through hard mask to realize top electrode of capacitive structure. PI substrate is released from the carrier wafer and Fig. 1 shows the process flow and fabricated devices. To make electrical connection, wire was attached on top and bottom electrode using silver paste and secured with epoxy glue.

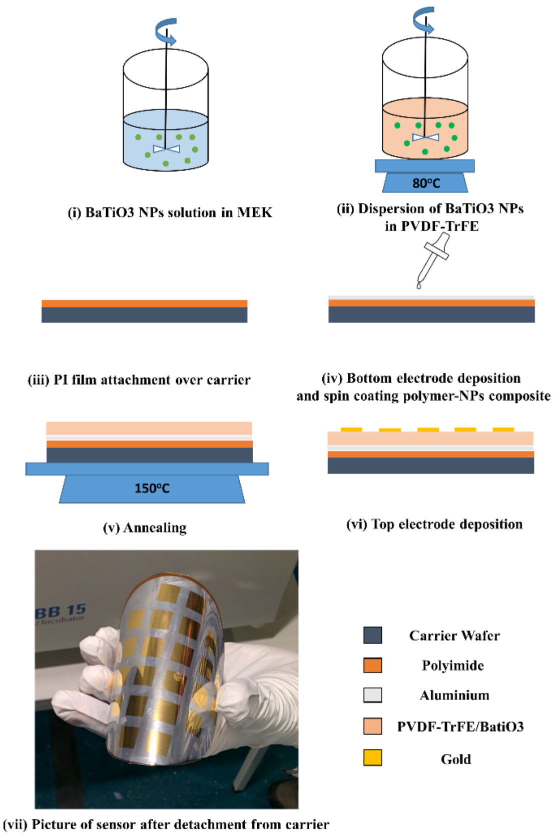

Figure 1: Process flow adopted for sensor fabrication

## III. CHARACTERIZATION

### *A. Composite Surface and Structure Chracterization*

To characterize the composite, we spin coated it on glass slide and annealed it at 150ºC. Atomic Force Microscopy (AFM) in tapping mode and Raman Spectroscopy were carried out to study the surface morphology and structural properties of material. AFM scan (Fig.2) reveals that, the addition of $BaTiO_3$ nanoparticles helps in better crystallization by providing nucleation site. During spin coating process, the nanoparticles are encapsulated by the polymer, and during annealing, the nucleation of the PVDF-TrFE occurs on the negatively charged surface of the $BaTiO_3$. Raman spectroscopy is carried out using LabRAM HR system with range of 200 $cm^{-1}$ to 1500 $cm^{-1}$. The spectrum as shown in Fig.2 suggest that the pristine PVDF-TrFE is dominated by β phase with pronounced peak at ~839 $cm^{-1}$ and α and γ phase are present in small proportion with just shoulder peaks at 813 $cm^{-1}$ and 883 $cm^{-1}$. Whereas in case of composite, the peak suppression is observed but the dominance of β phase is maintained, which signifies that the addition of $BaTiO_3$ favours the formation of β phase and reduces the α and γ content

### *B. Pressure Sensing*

The top and bottom electrode of sensor were connected to LCR meter for monitoring change in capacitance on application of force. The zero force capacitance value is measured to be 5.6 nF which gives the dielectric constant of 25, that is 2.5 more than that of pure PVDF-TrFE. A constant

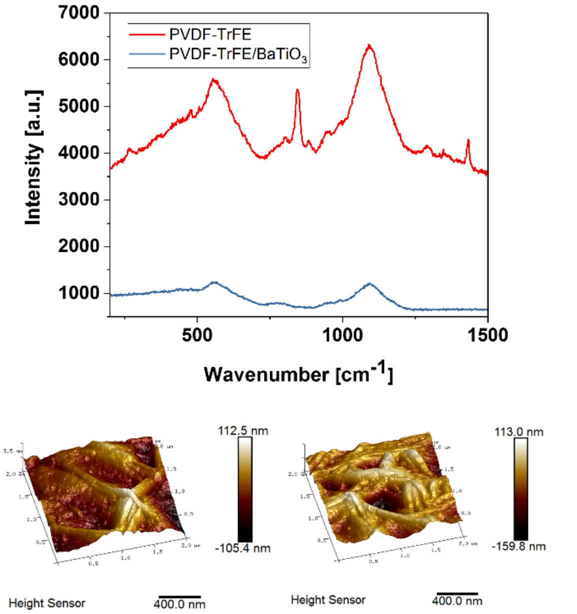

Figure 2: Raman spectroscopy and AFM scan image of pristine P(VDF-TrFE) (bottom right) and PVDF-TrFE/$BaTiO_3$ composite (bottom left)

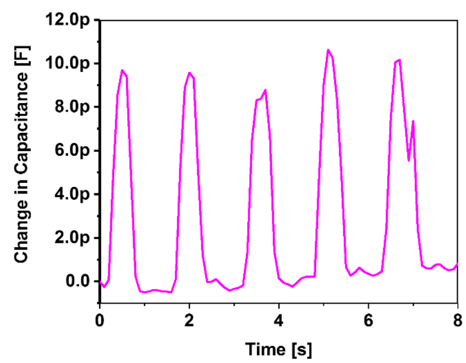

Figure 3: Response of sensor to periodic force of magnitude 45 mN

force of 45 mN was applied and released at regular interval of 2 secs to simulate a tapping form of touch. With every event of touch, ~10 pF increase in capacitance was observed which returns back to mean steady state value of around 5.6 nF upon release as can be seen from Fig.3. The slight upward drift in steady state value maybe due to relaxation time associated with transducer. Since, PVDF-TrFE is semi-crystalline polymer, change in thickness can be neglected and it can be said that the change in capacitance is due to piezoelectric effect of composite. During the characterisation, voltage across the capacitor was 1 volts, so change of 10 pF can be directly attributed to charge generation due to piezoelectric effect, which amounts to ~ 10pC. From this, we can calculate the piezoelectric coefficient as $d_{33} = Q/F$ and the value is 222 pC/N, which is much higher than the $d_{33}$ value of PVDF-TrFE (10-20 pC/N). This increase in piezoelectric coefficient value reflects that the addition of $BaTiO_3$ nanoparticles enhances the piezoelectricity of composite.

### *C. Temperature Sensing*

The sample was placed on hotplate and temperature is gradually increased. The room temperature was 27ºC and the initial capacitance at this temperature was measured to be 5.6

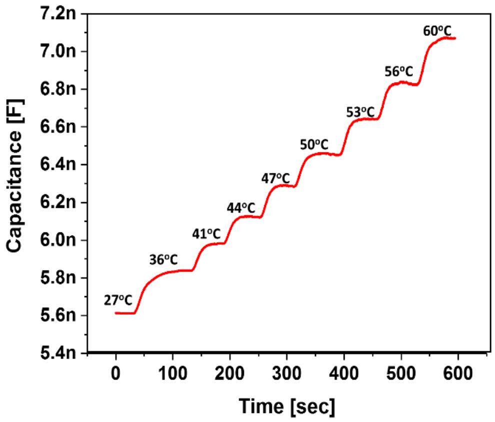

Figure 4: Response of sensor to stepwise increase in temperature

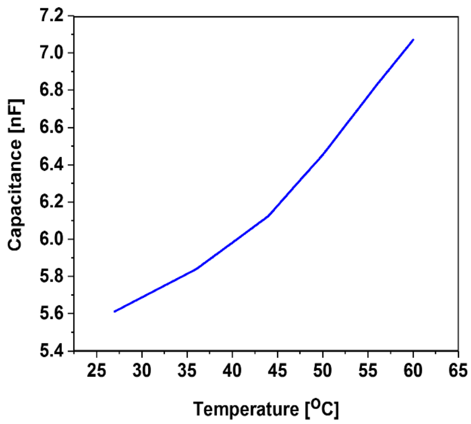

Figure 5: Plot of Temperature Vs Capacitance showing and the 54pF/°C sensitivity.

nF. With increase in temperature, the increase in capacitance is measured as shown in Fig.4. The measurement was done upto 60°C which is countered as high temperature in daily life activities and sensor shows almost a linear trend with sensitivity upto 54pF/°C as shown in Fig.5 which is 3.5 times more than that of pure PVDF-TrFE based temperature sensor reported in [12]. The increase in capacitance is attributed to increase in composite permittivity. Since dielectric permittivity of any material depends on electronics, ionic, dipolar and interfacial polarizations. Among these, effect of temperature on ionic and electronic polarization is very small, so increase in permittivity is directly related to increase in interfacial polarization of $BaTiO_3$ due to creation of crystal defects and decrease in dipolar polarization due to increase in randomness of dipoles.

## IV. CONCLUSION

The need of efficient and flexible sensor for E-Skin application has kept the researchers around the globe to come up with novel and high performance sensors. Therefore, a single sensor which can detect both touch and temperature is presented in this work using PVDF-TrFE-$BaTiO_3$ composite. The fabrication process is simple and can be extended to large area. Surface morphology and Raman analysis indicates that the addition of $BaTiO_3$ enhances the crystallization during annealing however, β phase formation decreases but remains in majority. The composite shows high sensitivity to temperature s (~54pF/°C) and pressure (0.22nF/N). Morover, due to an order of magnitude difference between the two sensitives, it can distinguish between the stimuli. The addition of $BaTiO_3$ nanoparticles in PVDF-TrFE on one hand gives a high dielectric constant material which can be used as energy harvester and on the other hand it increases the sensitivity of sensor manifolds. The fact the $BaTiO_3$ is lead free and PVDF-TrFE is flexible material, makes their composite a very strong candidate for sensing application.

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