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All-Fiber Pyro- and Piezo-electric Nanogenerator for IoT Based Self-Powered Health-Care Monitoring

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In this work, an all-fiber pyro- and piezo-electric nanogenerator (PPNG) is designed by multiwall carbon nanotube (MWCNT) doped poly(vinylidene fluoride) (PVDF) electrospun nanofibers as the active layer and interlocked conducting micro-fiber based electrode for converting both thermal and mechanical energies into useful electrical power. The PPNG generates high electrical throughput (output voltage $\sim 35$ V, maximum power density $\sim 34 \mu$W.cm$^{-2}$ and power conversion efficiency ($\eta_{\text{piezo}}$) $\sim 19.3\%$) with ultra-fast response time of $\sim 10$ ms. Owing to the higher piezoelectric charge co-efficient ($d_{33}/\sim 51.3 \text{pC/N}$) and figure of merit ($FoM \approx 5.95 \times 10^{-11} \text{Pa}^{-1}$) of PVDF-MWCNT nanofibers in comparison to the neat PVDF nanofibers ($d_{33}/\sim 22 \text{pC/N}$ and $FoM \approx 9.7 \times 10^{-12} \text{Pa}^{-1}$) the PPNG operates a range of consumer electronics component such as, capacitors and light emitting diodes. Furthermore, the electroactive phase content ($\sim 87\%$) is improved in the active layer due to the interfacial interaction between the surface charges from $\pi$-electron cloud of MWCNT and $-\text{CH}_2-$ dipoles of PVDF chain. Additionally, the PVDF-MWCNT nanofibers possess fifteen times higher pyroelectric coefficient ($\sim 60 \text{nCm}^{-2}\text{K}^{-1}$) than that of neat PVDF nanofiber ($4 \text{nCm}^{-2}\text{K}^{-1}$). As a result, the PPNG is capable to convert very large temperature fluctuations ($\Delta T \sim 14.30$ K) to electrical energy (such as, the open-circuit voltage of $250$ mV and a short-circuit current of $83$ pA). Besides this, it is capable to detect very low-level thermal fluctuations (as low as, $\Delta T \sim 5.4$ K) with responsivity ($\sim 1.48$ s) and possesses very high mechano-sensitivity ($\sim 7.5$ V/kPa) that makes it feasible to use as a biomedical sensor since the body temperature and bio-mechanical signals (such as breathing temperature, pulse rate, vocal cords vibrations, coughing sound, and so on) have an immense signature of health conditions. As a proof-of-concept, the all-fiber PPNG is employed as a biomedical sensor.
integrated with the Internet of Things (IoT) based human health care monitoring system as well as remote care of infectious disease (e.g., applicable for pneumonia, COVID-19) by transferring the pulse response, body temperature, coughing and laughing response wirelessly to a smartphone.

**KEYWORDS**: PVDF, MWCNT, electroactive phases, COVID-19, mechanical and thermal energy harvesting, nanogenerator, electrospinning, wireless sensor, IoT.
1. Introduction

Recently, “Internet of Things (IoT)” gained considerable attention in numerous applications, in remote health care monitoring systems, specially in infectious disease (e.g. pneumonia, COVID-19).\(^1\),\(^2\),\(^3\) An ultra-sensitive pressure-temperature dual functional pressure sensor is the main component of the stat-of-the-art IoT based remote health care system. It is used to collect the physiological signals from different parts of our body that behave as a transducer by converting an applied force and temperature fluctuations into an electrical signal, or other perceived signal output.\(^4\),\(^5\) These sensors can be adhered into our regularly used textiles or may be attached on the human skin for real-time monitoring of human health, activities and physiological signal measurements. In real-life application the textile based self-powered sensor is an ideal candidate as it is ultra-flexible, light weight and amenable to any surface of the body as well as no external bias is required to operate it. In this scenario, piezoelectric and pyroelectric materials are ideal candidate as it required no external bias to operate. Conventionally, very highly sensitive pressure sensors are witnessed by piezo-resistive and capacitive mechanism which needs external bias electric field. This essential requirement restricts the convenient utilization and integration of the sensors with the wireless communications systems. In terms of mechanical energy harvesting approach triboelectric nanogenerators are ideal candidate for higher power output generations\(^6\), but piezoelectric nanogenerators are essential device for higher stability and lower pressure sensing application, specifically physiological signal monitoring. To date, despite of brittle (low toughness) and rigid (high stiffness) in nature, a large number of inorganic and lead based materials have been applied to fabricate piezo- and pyro-electric nanogenerators.\(^7\) In contrast the organic polymer based piezoelectric and pyroelectric materials are of particular interest towards electronic skin because of their high compliance and flexibility, light weight nature and bio-compatibility. PVDF is one of the most favorable ferroelectric polymer classes.\(^8\)
Among the five crystallographic forms (α-, β-, γ-, δ- and ε), the β-phase is most suitable form for piezoelectric and pyroelectric properties of PVDF. There are several well-known methods available to attain high β-phase content, including the electrical poling method, where high electric field (typically 100 kV/mm) is applied. The electrical poling strategies can be overcome using the electrospinning method where the introduction of the β-phase and the *in situ* alignment of dipoles occur concurrently due to the nano-confinement effect in electrospun nanofiber. In addition, the electrospinning process enables the preparation of flexible and ultra-light piezoelectric membranes, making them more applicable in flexible self-powered sensors for human health detection or monitoring. However, the performance of electrospun PVDF nanofiber remains relatively low due to the lack of suitable device engineering, which leads to a small output power when used in energy harvesting applications. In this scenario, a large number of researchers have used different types of filler, especially; MWCNTs to improve the performance or efficiency of PVDF electrospun fiber based nanogenerators. The addition of MWCNTs to the PVDF significantly improved output responses for nanogenerators. From a materials point of view, the feasibility of MWCNTs as a filler material lies in its structural appearance, which made up of different coaxial cylinders composed of a single layer of graphene around the cylindrical inner hole. It is noticed that a huge numbers of articles are reported on PVDF nanofibers based nanogenerators, but temperature-pressure dual functional all-fiber based wearable sensor was hardly reported. The metal foil or metal-coated thin film-based electrodes for charge collection both are having limitations in the device lifetime under the prolonged cyclic stress. There were few more limitations found which includes the poor fatigue resistance causes early failure of the metal foils, huge mismatch of Young's modulus of the metal coating electrode and the active thin film. However, due to prolonged period of application, loss of mechanical integrity and electric connectivity occurred. Now all these issues are being
avoided from the present state of devices by adopting an efficient and durable all-fiber nanogenerator of a three-layered structure where both piezo- and pyro-electric active constituent and electrodes are composed of adaptable and soft fiber arrays.

In this work, a notable enhancement of electroactive phase content in electrospun PVDF nanofiber has been attained by synergistic effect of interfacial interactions of MWCNTs with PVDF chain and further mechanical stretching in the course of collection of fiber by high speed rotating collector. As a result very high piezoelectric/pyroelectric coefficient, figure of merit and energy harvesting output power with high power conversion efficiency has been achieved. A highly sensitive all-fiber nanogenerator is designed with PVDF-MWCNTs nanofibers which could capture very low level temperature fluctuation and possesses very high pressure sensitivity. With such effective performance, an IoT based wireless health care system is developed through which the physiological signals are possible to transfer to the smart phone, indicating a promising way of real time remote health care monitoring.

2. Results and discussion:

All-fiber Nanogenerator/Sensor Fabrication Strategy:

The copper (Cu)–nickel (Ni) plated interlocked micro-fiber based polyester fabric was used as top and bottom electrodes to fabricate the piezoelectric and pyroelectric nanogenerator (PPNG). Conducting copper wires were attached on both electrodes and finally the three layered structure PPNG was covered with a PDMS layer. The schematic diagram of the all-fiber nanogenerator is shown in Fig. 1a. The generated electrospun nanofibers are shown in Fig. 1a(i). The all-fiber nanogenerator consists of PVDF-MWCNT electrospun nanofibers (Fig. 1a(ii)) sandwiched between interlocking micro-fiber arrays of conducting fabric as charge collecting electrodes (Fig. 1a(iii)). The device was further encapsulated with PDMS for protection from environment and to ensure compact structure of the device. The diameter of the interlocked micro-fiber of the conducting electrode was of 15 μm. The excellent
flexibility and conformability of the generated electrospun nanofibers to human finger is shown in the Fig. 1b. The digital image of fabricated original PPNG (shown in Fig. S1, ESI). Thus, the PPNG fabricated by the active piezoelectric and pyroelectric component with the electrodes, all are composed of fibers.

**Structural and Electrical Properties**

The FE-SEM image with the corresponding histogram profiles of PVDF-MWCNT nanofibers is shown in Fig. 2a. The fiber diameter distribution plot is presented in the inset of Fig. 2a. It was observed that the randomly oriented nanofibers are formed and the average diameters of the fibers are ranging from 40-165 nm, while the maximum number of fibers possesses a diameter of ~ 85 nm. During the formation of fibers i.e., electrospinning process, the molecular dipoles of PVDF nanofibers are preferentially oriented in the out-of-plane direction since the nanofibers and polymer chains are aligned within the plane of the collector. This is due to the simultaneous application of electric field and mechanical stretching leading to polymer jet elongation, whipping and *in situ* poling. During electrospinning of the MWCNTs mixed PVDF solution, the conductive MWCNTs can produce inductive charges on its surface when the external electric field is applied, which results to a greater Coulomb force during the electrospinning process. This effect helps to improve piezoelectric and pyroelectric properties of electrospun PVDF-MWCNTs nanofibers due to excellent interfacial interaction between the PVDF polymer chain and MWCNT surface charges at nanoscopic scale. Therefore, a synergistic effect occurred between interfacial interaction and mechanical stretching during collection of fiber by high speed rotating collector during crystallization of fibers. As a result, the content of electroactive β-phase was much higher in PVDF-MWCNT nanofibers than that of the neat (pure) PVDF nanofibers and this is evident from the FT-IR plot, see Fig. 2b. FT-IR spectra of PVDF-MWCNT and neat PVDF nanofibers are shown in the Fig. 2b, in the region of 1600–400 cm$^{-1}$.
Evidently the neat PVDF nanofibers contain the α-phase (non-polar) (see vibrational band at 762 cm⁻¹ in the inset of Fig. 2b and β-, γ-(polar) phases (vibrational band at 1277 and 1234 cm⁻¹ respectively). However, the PVDF-MWCNT nanofibers contain only polar β- and γ-phases and no α-phase peak were available in spectrum, as seen in the inset of Fig. 2b. This implies that the conversion of non-polar phases (α-phase) into polar phases (β-, γ-phases) in PVDF-MWCNT nanofibers in comparison to neat PVDF nanofibers by electrospinning process. Eventually, it is found that both the nanofibers having 841 cm⁻¹ vibrational band, which presenting the β- and γ-phases concurrently.

Assuming that the infrared absorption follows the Beer-Lambert law and relative proportion of electroactive β-, γ-phases phases (defined as \( F_{EA} \)) is calculated using equation (1) as,

\[
F_{EA} = \frac{A_{841}}{(K_{762})A_{762} + A_{841}} \times 100\%
\]

where, \( A_{762} \) and \( A_{841} \) represent the absorbencies at 762 and 841 cm⁻¹ respectively, \( K_{762} = 6.1 \times 10^4 \) cm²mol⁻¹ and \( K_{841} = 7.7 \times 10^4 \) cm²mol⁻¹ representing the absorbance coefficient corresponding to wave numbers. Therefore, the calculated \( F_{EA} \) of neat PVDF nanofibers is \( F_{EA} \sim 67\% \), whereas in the case of PVDF-MWCNT nanofibers, \( F_{EA} \sim 87\% \).

Additionally, Fig. 2c shows the curve-deconvoluted XRD patterns of the PVDF-MWCNT and Neat PVDF nanofibers. As evident, the diffraction peak at 20 of 19.2° in neat PVDF nanofibers corresponds to the reflection of α-crystalline phase (110) and 20 of 20.8° corresponds to the cumulative peak of β- and γ- crystalline phase (110/200). Although electrospinning technique is a very good option to induce all-trans configuration (β-phase) in the PVDF crystal structure, the α-phase still exist in the neat PVDF nanofibers that can be understood from the XRD and FT-IR analysis. The α-crystalline phase is fully contracted in the composite fibers (Fig. 2b and Fig. 2c), which is due to the addition of MWCNT in the PVDF matrix. The conversion of non-polar to polar phase is carried way due to two factors:
1. Simultaneous poling and stretching during the electrospinning process.

2. The interfacial interaction present between the surface charge /π-cloud of MWCNT and –CH\_2– dipoles of PVDF chain.

The degree of crystallinity (χ\_c) is calculated from \( \chi_c = \frac{\sum A_{cr}}{\sum A_{cr} + \sum A_{amr}} \times 100\% \) (2), where \( \sum A_{cr} \) and \( \sum A_{amr} \) are the summation of integral area of crystalline peaks and amorphous halo respectively. Due to addition of MWCNT, the total crystallinity of PVDF-MWCNT nanofibers is higher (59%) than that of neat PVDF (53%). The crystallization along with thermal stabilization of β-phase in PVDF-MWCNTs was evaluated by differential scanning calorimetry (DSC) study, as shown in Fig. 2d.

It can be seen that both the melting peaks (t\_m) of two types of nanofibers indicate the existence of β- and γ-phases. The endothermic melting peak of the β- and γ-phases is found in PVDF-MWCNT nanofibers at (~168 °C) and (~179 °C) respectively.\(^{16, 19-21}\) For the neat PVDF nanofibers, it is found that the melting peak of α/β-phases is (~160 °C).\(^{16, 19-21}\) As a result, both the β- and γ-phases are considered more stable thermally in the PVDF-MWCNT nanofibers with respect to neat PVDF nanofibers. As a consequence, the crystallization temperature (t\_c) of PVDF-MWCNT nanofibers (~148 °C) was also enhanced with respect to the neat PVDF (~141 °C). This is due to the MWCNTs acting as a nucleating agent and hinders the movement of polymer chain segments. Moreover, the induced polar β-phase is stabilised during rapid crystallization and not relaxed back to non-polar α-phase via thermal motion. As a result, the formation of the crystalline β-phase is irreversible in the PVDF-MWCNT nanofibers and not relaxed back to α-phase via thermal motion.\(^{22}\) Noteworthy that in the case of free standing film sample containing of gamma phase, the melting points shows relatively much higher side.\(^{19}\) In contrast, electrospun samples act a bit differently it may be because of air-permeable spongy like behaviour that leading to lower melting temperature.
than the right film specimen. In addition, from the DSC data the total degree of crystallinity ($\chi_{ct}$) of the nanofibers was calculated using, 

$$\chi_{ct} = \frac{\Delta H_m}{(1 - \varphi)\Delta H_m^0}$$

where, $\Delta H_m$ represents the melting enthalpy, $\Delta H_m^0$ is the melting enthalpy of the 100 % crystalline PVDF ($\sim 104.7 \text{ Jg}^{-1}$) and $\varphi$ is the weight fraction of MWCNT. The value of $\chi_{ct} = 40\%$ and 33% for PVDF-MWCNT and neat PVDF nanofibers respectively. In overall, the origin of improved electroactive phase content and improved degree of crystallinity and higher thermal stabilization is the synergistic effect. Therefore, it is concluded that the degree of crystallinity of PVDF-MWCNT nanofibers is improved due to the presence of MWCNT.

From materials point of view, it is very evident that PVDF-MWCNTs nanofibers are proven to be an excellent material for the application of piezo- and pyro-electric nanogenerator. 

of particular interest, interfacial interaction between the MWCNT NPs and PVDF dipoles is schematically shown in Fig. 2e. Here, MWCNT mainly acts as a nucleating agent. During the electrospinning process, charge is accumulated at the interfacial boundaries between MWCNT and non-polar $\alpha$- phase and ferroelectric $\beta$- phase. In addition, there might be present dipolar intermolecular interaction between $\pi$-cloud of MWCNT and $-\text{CH}_2-$ dipoles of PVDF chain. So, inter-conversion of $\alpha$- to $\beta$- phase near the MWCNT surface can be described by the charge accumulation at the interphase and dipolar interaction. Moreover, the induced polar $\beta$- phase is stabilised during rapid crystallization and not relaxed back to non-polar $\alpha$-phase via any thermal motion.

**Pyroelectric Energy Harvesting**

The harvesting of thermal energy from the all-fiber PPNG is obtained by pyroelectric effect. The temperature fluctuations is maintained using an IR light source placed on the top surface of the PPNG. The pyroelectric energy harvesting performance of the PPNG is shown in Fig. 3 in terms of the measurement of open circuit voltage ($V_{oc}$) under different temperature fluctuations of $\Delta T \sim 14.3 \text{ K}$ (Fig. 3a), $\Delta T \sim 5.4 \text{ K}$ (Fig. 3c) as well as corresponding short
circuited current ($I_{sc}$) with the temperature gradient ($dT/dt$) in Fig. 3b and Fig. 3d respectively. It is noteworthy to mention that the $I_{sc}$ and $V_{oc}$ are 71 pA and 250 mV (approx.) respectively when $\Delta T \sim 14.3$ K and $dT/dt= 2.15$ Ks$^{-1}$ under the temperature switching frequency of 0.01 Hz which is much higher than the device made from neat PVDF. When the switching frequency changes from 0.01 Hz to 0.1 Hz ($\Delta T \sim 5.4$ K and $dT/dt= 1.72$ Ks$^{-1}$) the $I_{sc}$ and $V_{oc}$ are 83 pA and 135 mV (approx.) respectively as shown in the Fig. 3c-d. The short circuited current $I_{sc}$ can be expressed as $I_{sc}= A \frac{d\rho dT}{dtdT} = Ap \frac{dT}{dt}$ (4), where $A$ is the surface area (m$^2$) and $\frac{dT}{dt}$ is the rate of temperature change (K/s) and $\rho$ is the pyroelectric coefficient (nCm$^{-2}$K$^{-1}$). This enhancement of $I_{sc}$ was accomplished by a thermally induced piezoelectric coupling effect, as the output performance is proportional to $\Delta T$ and $dT/dt$. The available maximum power density ($P$) of the PPNG is estimated to be $\sim 12$nWm$^{-2}$ when temperature difference between the electrodes exist $\Delta T \sim 14.3$ K. Using equation 4, the estimated pyroelectric coefficient ($\rho$) of the PPNG is 60 nCm$^{-2}$K$^{-1}$, which is almost fifteen times higher than the neat PVDF fiber (4 nCm$^{-2}$K$^{-1}$). The $I_{sc}$ and $V_{oc}$ are found to be linearly proportional to $dT/dt$ and $\Delta T$ profiles, respectively (Fig. 3e,f) in reliability with theoretical paradigm. These linear characteristics suggest the potential of the PPNG as a self-powered pyroelectric temperature sensor for future practical applications especially in various infectious diseases. Fig. 3g shows the expanded view of positive current pulse signal of the rectangular red dotted marked cycle of Fig. 3b. It has been observed that the current increases from zero to its peak value within $\sim 1.48$ s at positive side while in negative side it takes $\sim 1.54$ s (inset of Fig. 3g). As soon as the heat source is removed, the positive output current is returned back to a minimum value. Fig. 3h represents the “$I_{sc}$ vs $T$” curve where it is observed that the current pulse is decreasing exponentially with a time constant value of 2.60 s. This decaying process corresponds to a temperature reduction of the PPNG. The value of the reset time for the PPNG is estimated as the time required recovering to $1/e$ or (37%) and
is approximately 2.60 s. So, it is noteworthy to mention that the PPNG can be used as a good temperature sensor due to its reduced response time and reset time than the previously reported thermoelectric sensor.}

The better pyroelectric performance in PVDF-MWCNT nanofibers is ascribed to the infrared transparency (~ 5%) and improve absorption property of electromagnetic energy by MWCNT which generated rapid heating in the nanofibers. When the device is exposed under heat (i.e. $dT/dt > 0$) the short circuited current ($I_{sc}$) gets its positive peak and a negative peak through the cooling process (i.e $dT/dt < 0$). This is happened because of the decrement of the degree of spontaneous polarization at the time of heating process, which forces the surface charges to flow within the two polar surfaces. The current flows in the opposite direction because of the attractive force of the free charges to the polar surfaces when the device is subsequently cooled. From the output voltage and current graph, it is very evident that output current follows the temperature gradient profile (as, $I_{sc} \propto dT/dt$) and output voltage follows the thermal fluctuations ($V_{oc} \propto AT$) profiles. A summary or comparison of device materials, electrode materials, output performance and pyroelectric coefficient of the PPNG with the reported pyroelectric nanogenerators is shown in Table S1 (ESI). It has been observed that in spite of lower pyroelectric co-efficient of PVDF-MWCNT nanofibers in many cases the output performance, in terms of output voltage and current is much higher than that of previous reports.

**Piezoelectric Energy Harvesting**

To demonstrate the application of the materials, we have fabricated a PVDF-MWCNT based all-fiber nanogenerator/energy harvester (PPNG). Electrical characterization of the PPNG is shown in Fig. 4a-f and repetitive finger striking over the PPNG surface from a 5cm height is undertaken with different magnitude of compressive stress ($\sigma_a=2$, 4 and 6 kPa), as shown in the Fig. 4a. As a result, the PPNG generates open circuit output voltages ($V_{oc}$) of 11, 20 and
35 V respectively which shows more than 90% improvement in output voltage under magnitude of compressive stress $\sigma_a=2$ kPa over the neat PVDF based nanogenerator (see Fig. S2, ESI). In addition, the press and release response of single peak from PPNG is shown in the right lower inset of Fig. 4a and it is found that the PPNG shows ultra-fast response time of $\sim 10$ ms. It is important to note that generated $V_{oc}$ varies almost linearly with the increase of $\sigma_a$ (right upper inset of Fig. 4a) which is consistent with the piezoelectric theory. As a result, the mechanosensitivity ($S_M$), where $S_M= (\Delta V_{oc} / \Delta F)$, has been determined to quantify the dynamic mechanical stimuli sensing ability where $\Delta V_{oc}$ and $\Delta F$ are the differences of the output voltage and force respectively.\textsuperscript{12,22} The average mechano-sensitivity ($S_M$) of the PPNG is $\sim 7.5$ V/kPa, which is higher than the earlier published high performance nanogenerators.\textsuperscript{18,24,29-31} A detailed comparison plot of piezoelectric mechano-sensitivity with cited references is provided in Fig. 4b. The horizontal line use to guide the eyes whereas vertical line indicates improved value of this work. It is very prominent that sensitivity of our device is much improved over previous research and very much suitable for healthcare monitoring applications where sensitivity is the major factor. The superior energy harvesting performance of the PPNG is attributed to the higher piezoelectric charge coefficient $d_{33} = g_{33} \varepsilon_0 \varepsilon_r = 51.3 \text{ pC} / \text{N}$, where $g_{33}$ is piezoelectric voltage coefficient, $\varepsilon_0$ is permittivity of free space ($\sim 8.85 \times 10^{-12} \text{ F m}^{-1}$) and $\varepsilon_r \approx 5$ at 1 KHz is dielectric constant of PVDF-MWCNT nanofibres.\textsuperscript{38,39} The estimated piezoelectric voltage coefficient was found as, $g_{33} \approx \frac{V_{oc}}{\sigma_a \times t} = 1.16 \text{ Vm/N}$, where $\sigma_a= 6$ kPa is stress magnitude and $t$ is the thickness of PPNG.\textsuperscript{39} Thus, PVDF-MWCNT exhibits a superior piezoelectric figure of merit ($FoM \approx g_{33} d_{33}$ $\approx 5.95 \times 10^{-11} \text{ Pa}^{-1}$) than those of the neat PVDF nanofibers ($d_{33} \approx 22 \text{ pC} / \text{N}$ and $FoM \approx 9.7 \times 10^{-12} \text{ Pa}^{-1}$) and other various state-of-the-art PVDF-based nanofibres.\textsuperscript{22} The improvement in piezoelectric coefficient with the addition of MWCNTs is due to high
amount of β-phase in the PVDF. We propose that the presence of MWCNTs alters the orientation of the dipoles in the PVDF nanofibers. The incorporation of fillers causes PVDF to swell leading to the formation of extended chain conformation of PVDF around the fillers. Furthermore, adding MWCNTs to reinforce PVDF fibers can enhance the crystallinity of the β-phase enhancing piezoelectric properties. The $d_{33}$ value of the PVDF-MWCNT nanofibers is found to be significantly greater than those of various PVDF-based nanofibres, a comparison of nanofibres is shown in Table S2 (ESI). To explore the PPNG as an energy harvesting power source, we have demonstrated in some other way where the output voltages were measured as a function of the external load resistors ($R_L$) (Fig. 4c) and it is obvious that the instantaneous output voltage drop across the resistive load steadily builds up as the load resistance increases and remains constant at infinitely large resistance (4 MΩ). In addition, in the same figure (Fig. 4c) the variation of the load current with the external load resistors is shown. The instantaneous output power density ($P$) of the PPNG was calculated as $P = \frac{1}{A} \frac{V_L^2}{R_L}$

\[ \text{……… (5)} \]

where $A$ is the effective contact area, and $V_L$ is the voltage drop across load resistance $R_L$. It is obvious from the Fig. 4d that the available instantaneous output Power density ($P$) of 34 μW.cm$^{-2}$ at the load resistance ($R_L$) of 3.4 MΩ has been accomplished and it is noted that this high output power density to be enough to make on few (~10) blue LEDs when subject to a mechanical pressure without using a storage system. In the interest of demonstrating a practical application of the all-fiber PPNG, the PPNG was connected to external capacitors (1μF, 2.2μF and 4.7μF) through a full-wave bridge rectifier circuit and charging up under periodic human finger imparting stress amplitude 6 kPa and capacitor charging behavior is shown in Fig. 4e. It is important to note that PPNG was able to charge up capacitors of several capacitances (such as, 1μF, 2.2μF and 4.7μF) to 1.18 V, 1.07 V and 0.65 V in 55 sec, 60 sec and 78 sec respectively. The value of the time constant ($\tau$) for all the capacitors are calculated as the time required recovering to (1-1/e) or (63%) of its steady state
voltage are approximately 19 s, 20 s and 48 s respectively that ensures the rapid energy supply ability of the PPNG. The power (P) stored in the capacitor is calculated as,  

\[ P = \frac{CV_s^2}{2t} \] 

…..(6), where C is the capacitance of the charging capacitor, \( V_s \) is the saturation voltage, and t is the time required to reach the saturation voltage. The estimated powers stored in the capacitors are found to be 0.013 \( \mu \)W, 0.021 \( \mu \)W and 0.013 \( \mu \)W respectively. Fig. 4f has shown the rectified \( V_{oc} \) under 6 kPa of compressive stress using a bridge rectifier circuit (right upper inset) which was used to charge up the capacitors. Larger is the deformation in the nanofibers when the applied compressive stress is higher and as a result enhancement of piezo- potential is found in the PPNG. More importantly, the long term stability in the output voltage generation from the PPNG is shown in Fig. S3,ESI.

The instantaneous piezoelectric energy conversion efficiency (\( \eta_{piezo} \)) of PPNG is defined as the ratio of the generated output electrical energy (by the instantaneous compressive axial stress and the residual mechanical vibration after the impacting) \( (E_{elec}) \) to the input mechanical energy \( (E_{mec}) \). The optimum resistance (3.4 M\( \Omega \)) was connected with the PPNG to derive optimized output performance and the output voltage per cycle was measured instantaneously, as shown in Fig. 4c and Fig. S4.

\[ E_{elec} = \int_0^t \frac{V(t)^2}{R} dt \] 

\( \ldots . . . . . . (7) \),

The evaluated output energy, \( E_{elec} = 8.33 \times 10^{-6} \) J.

To determine the input mechanical energy \( (E_{mec}) \) we have to consider the total axial deformation of the PPNG under 6 kPa stress as, \( \Delta L = \varepsilon \times L \). Here, \( \varepsilon \) is the developed axial strain and \( L \) is the total thickness of the device. The generated axial strain is given by, \( \varepsilon = \frac{\sigma_a}{Y} \)

\( = 0.13 \) where \( Y \sim 0.31 \) GPa is the Young’s modulus of the PVDF-MWCNT nanofibres.40
Thus, the total input mechanical energy per cycle is calculated by, $E_{mec} = F \times \Delta L = 0.63 \times 10^{-6} \text{ J}$ where $F$ is the applied force.

The instantaneous piezoelectric energy conversion efficiency of the PPNG can now be written as, $\eta_{\text{piezo}} = \frac{E_{\text{elec}}}{E_{\text{mec}}} \times 100 = 19.3\%$.

Finally it is found that the instantaneous piezoelectric energy conversion efficiency of the PPNG is superior to that of previously reported works.\textsuperscript{22,41} A comparison of device materials, electrode materials and percentage of piezoelectric energy conversion efficiency ($\% \eta_{\text{piezo}}$) of the PPNG with the reported nanogenerators is shown in Table S3, (ESI) where in many cases the $\% \eta_{\text{piezo}}$ of our developed device is much higher than that of previous works.

A summary or comparison of device materials, electrode materials, and output performance of the PPNG with the reported nanogenerators is shown in Table S4, (ESI) where in many cases the output power of our developed device is much higher than that of previous works. Importantly, previous research on MWCNT reinforced PVDF composites didn't explore the temperature and pressure dual functionality in the device form which is the one of main focus of this research. Table S5, (ESI) clearly indicates the uniqueness of this work in terms of both thermal and mechanical energy harvesting over previous research.

**Healthcare Monitoring System**

In this section PPNG is being used as a self-powered sensor. Here PPNG was attached on the human skin to detect slight movement of the palm, wrist, arm muscle, leg muscle, throat (coughing and laughing situation) and different voltage responses are shown in the Fig. 5a-f and corresponding digital picture during the test is shown in the inset. The responding positive and negative voltages generated by palm and wrist bending and releasing are shown in Fig. 5a,b which are mainly caused by the deformation of epidermis during wrist
movements. Fig. 5c,d are shown the output voltage response to the arm and leg muscle movement. Furthermore, the PPNG could transduce the vocal cord vibrations (Fig. 5e,f) to the coughing and laughing situation when the device is attached to the throat. Fig. 5g,h presented the Fast Fourier Transform (FFT) of the coughing and laughing signal (shown in Fig. 5e,f). The FFT treated cough acoustic frequency spectrum 200-380 Hz and laughing acoustic frequency spectrum 126-424 Hz is presented respectively which is good agreement with the healthy people coughing and laughing sound.\textsuperscript{22, 42-43} Therefore, the PPNG can be used as self-powered sensor to monitor and separate virus infected patients from normal humans. It is noteworthy to mention that the PPNG can be used for self-powered human-motion detection as it exhibited a good repeatability and high responsivity to strain variation caused by the joint/muscle movements without external power supply. Also this sensory information from skin attachable/wearable PPNG is beneficial for body movement analysis during sports activities and shows the potential of using as epidermal device for skin motion monitoring.\textsuperscript{44}

**Remote Health Monitoring**

Here we have demonstrated a real time practical application of remote health monitoring system which is very much important for continuous monitoring of both viruses suspected and infected patients reducing the risks of caregivers being exposed to the virus. We have designed it in two-way methods using i) Bluetooth Module ii) Nodemcu Wi-Fi Module. Fig. 6a shows the basic circuit diagram of the experimental setup for using a Bluetooth Module and Fig. 6b shows the basic circuit diagram of the experimental setup for using IoT based remote health care monitoring system. In the first case a Bluetooth Module (HC-05), one Arduino UNO board and an android mobile phone with Blynk app are the main components whereas in the second method, the main elements/components are single chip ESP8266A Wi-Fi module and a PC/Smart phone with Blynk app. The single chip ESP8266A has 512KB
Flash memory and 10 bit ADC in set. Its run on L106, 32 bit MCU with Real Time Operating System (RTOS). The clock speed is 80MHz. also it has a 802.11 b/g/n and frequency range is 2.4G-2.5G (2400M-2483.5M) system. With the help of MCU, we performed analog to digital conversion of PPNG/sensor data value for processing. The waveforms are processed and corresponding data are sent to a local server made by using ESP8266A HTTP protocol. The patient or doctor can easily check the report to access on this network through Wi-Fi/Bluetooth using their smart phones and laptops. Fig. 6c shows the real time practical circuit which shows the PPNG/sensor response under repeated finger touch in the smart phone screen through the local server for IoT based remote health care monitoring system and Fig. 6d shows the plotted sensor output graph.

Using this system clinician can monitor several physiological signals such as, heartbeat, respiration rate, body temperature, coughing signal and so on of the virus infectious/suspected patients even in quarantine situation round the clock wirelessly without the direct physical contact to the patient which simply prevent further spread of the virus. In future, envisioned strategy through non-invasive piezo- and pyro-electric-based wearable sensors will be taking place for infectious diseases and in vivo body implantable.\textsuperscript{45,46}

3. Conclusions

In summary, we have developed a single device platform by all-fiber nanogenerator for harvesting both the mechanical and thermal energy using PVDF-MWCNTs electrospun nanofibers as active piezo- and pyro-electric component respectively. It has been observed that the content of electroactive phases, degree of crystallinity and thermal stabilization of $\beta$-phase within PVDF nanofibers was mainly affected by the synergistic effect of interfacial interactions and mechanical stretching forces exerted during collection of fibers. As a consequence, mechanically robust structure of PVDF-MWCNT nanofibers possess enhanced piezoelectric charge co-efficient ($/d_{33}/$~51.3 $pC/N$) and figure of merit ($FoM \approx 5.95 \times$
10^{-11} Pa^{-1} ) of PVDF-MWCNT nanofibers in comparison to the neat PVDF nanofibers (\(d_{33}/\sim 22\text{ pC} /N\) and \(F_o M \approx 9.7 \times 10^{-12} Pa^{-1}\)). In addition to that the PPNG shows ultra-fast response time of \(\sim 10\text{ ms}\), high electrical throughput (output voltage \(\sim 35\text{ V}\) and maximum power density \(\sim 34\text{ µW.cm}^{-2}\)) and power conversion efficiency (\(\eta_{\text{piezo}}\sim 19.3\%\)).

As a result, the nanogenerator drives several consumer electronics components such as, commercial capacitors and LEDs by simple human finger imparting. In addition, we have also developed a human health care monitoring system where our developed sensor can distinguish different muscle vibration signal from different parts of our body. Finally, a wireless health care system for remote health monitoring using IoT and a Bluetooth module with an Arduino Uno is developed by which a clinician can monitor several physiological signals of the virus infected/suspected patients round the clock wirelessly without the direct physical contact to the patient which simply prevents further spread of the virus. This type of healthcare system is urgently needed for the frontline health staffs during current pandemic.

In overall, the integrated device design, portable feature, higher thermal and mechanical sensing capabilities of our developed all-fiber device could find the possible applications in artificial electronic skin, prosthetic limbs, rehabilitation programs and other artificial intelligence applications in near future to monitor dynamic tactile and strain information.

4. Experimental section

Materials

PVDF pellets (Mw=275 000, Sigma–Aldrich, USA), acetone (Merck Chemical, India), N, N-Dimethyl formamide (DMF from Merck) and MWCNT powder (Merck, India).

Remote health care system: Arduino Uno which is an ATMega 328 microcontroller based system, Arduino Uno based Wi-Fi Shield ESP8266MOD which is used as IoT based gateway for establishment of communication with IoT cloud and the system and a Bluetooth module (HC-05) and our proposed nanofiber based sensor.
Fabrication of electrospun nanofibers

The PVDF solution was prepared by dissolving 12wt% of PVDF pellets in DMF/acetone (60:40 v/v) solution under continuous stirring until solution was acquired. Then 0.2 wt% (w/v %) of MWCNT was added to the resulting solution. The solution was stirred, followed by sonication in an ultra-sonication bath at room temperature until uniformly dispersed MWCNTs within the PVDF solution was obtained. Then, the resulting solution was placed in a plastic syringe (15 mL) tipped with a stainless steel needle with an inner diameter of 0.5 mm. Electrospinning was carried out at 11 kV from a high voltage power supply, where stainless steel needle is connected to positive terminal of high voltage power supply and metallic collector is grounded. A syringe pump was applied to deliver the solution into the needle tip at a flow rate of 0.5ml/hr and the electrospun nanofibers were collected through aluminum-foil covered roller collector. The needle-to-roller collector distance of 12 cm was optimized. All experiments were supervised at room temperature and a relative humidity of 43 ± 5%. The schematic of electrospinning set up used for fabrication of large area PVDF-MWCNT nanofiber mat is shown in Fig. S5, ESI.

Fabrication of all fibers PPNG

The details of the fabrication process of our proposed all fibers PPNG has been discussed in section “All-Fiber Nanogenerator/Sensor Fabrication Strategy”, under results and discussion part. The detail of the device structure is shown in Fig. 1. Here the copper (Cu)–nickel (Ni) plated interlocked micro-fiber based polyester fabric (supplied by Coatex Industries, India) was used as top and bottom electrodes and PVDF-MWCNT nanofibers is used as an active layer. Finally the PPNG was further encapsulated with PDMS (Sylgard 184 silicone elastomer) for protection from environment and to ensure compact structure of the device. The complete thickness (t) of the PPNG is ~5 mm and effective contact area of the PPNG is 7.5 cm².
5. Characterization

To investigate in detail the surface morpholigc of the electrospun fibers, field emission scanning electron microscopy (FE-SEM, FEI, INSPECT F50) was undertaken at an acceleration voltage of 20 kV. The structural composition of the electrospun fibers were measured by Fourier transform infrared spectroscopy (FT-IR) (Bruker, Tensor II). X-Ray diffraction (XRD) patterns of the electrospun nanofibers were recorded in an X-Ray diffractometer (Bruker D8 Advance) with CuKα (λ ~ 1.5406 Å) monochromatic radiation operated at 40 kV and 30 mA. Finally the output voltages response from PPNG under different applied stress was recorded through a digital oscilloscope (Agilent DSO3102A) and current measurement was carried out with Keithley 6485 picoammeter.

ASSOCIATED CONTENT

Electronic Supporting Information

The digital image of fabricated PPNG, piezoelectric response for neat PVDF based nanogenerators, the schematic of electrospinning set up used for fabrication of large area PVDF-MWCNT nanofiber mat.

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Conflict of interest

The authors declare no competing financial interest.

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Figure 1: (a) Schematic of the all fiber PPNG comprising of (i) large area PVDF-MWCNT nanofiber mat (8 cm × 7 cm). (ii) FESEM images of PVDF-MWCNT nanofiber and (iii) micro-fiber based conducting electrode. (b) The excellent flexibility and conformability of PVDF-MWCNT electrospun nanofibers is shown by wrapping it on a human finger.
Figure 2: (a) FE-SEM image and corresponding fiber diameter distribution plot in the inset, (b) FT-IR spectra of PVDF-MWCNT and neat PVDF nanofibers in the region of 1600–400 cm⁻¹, (c) XRD patterns and their curve deconvolution of PVDF-MWCNT nanofibers and neat PVDF nanofibers, (d) DSC thermograms of the PVDF-MWCNT and neat PVDF nanofibers.
(e) Schematic of interaction between \(-\text{CH}_2\)- dipoles of PVDF chain and \(\pi\)-electron clouds/surface charges of MWCNT.
Figure 3: Measurement of output open circuit voltage under temperature fluctuations of (a) \( \Delta T \sim 14.3 \) K and (c) \( \Delta T \sim 5.4 \) K and corresponding output short circuit current with the variation of rate of temperature \( (dT/dt) \) in (b) and (d) respectively. (e) Variation in output current as a function of rate of change in temperature. (f) Output voltage as a function of temperature difference. (g) The expanded view of positive output current signal of the marked cycle in (b) and negative pulse signal in the inset. (h) The expanded view of current \( (I_{sc}) \) vs time \( (T) \) curve of the marked cycle in (b) when the source of heat was removed.
Figure 4: Energy harvesting characterizations of nanogenerators. (a) Pressure dependent output open circuit voltage ($V_{oc}$) of PPNG, (b) mechano-sensitivity of reported piezoelectric based pressure sensors. The abscissa indicates cited references. (c) the variation of output voltage and current with external load resistances, (d) the output power as a function of
external load resistances, (e) capacitor charging behavior with different capacitor values, (f) output open circuit rectified voltage response with bridge rectifier circuit.
Figure 5: PPNG for detecting human motions as a sensor. Open circuit voltages when PPNG is attached on human skin to detect (a) palm bending, (b) wrist motion, (c) arm muscle bending, (d) leg muscle bending, (e) coughing signal and (f) laughing signal. Insets show the corresponding photography images for the tests, (g) FFT treated cough acoustic frequency spectrum and (h) laughing acoustic frequency spectrum.

Figure 6: Remote Health care monitoring system, (a) circuit diagram of the Bluetooth Module based experimental setup; (b) circuit diagram of the IoT based experimental setup, (c) digital image of practical circuit with mobile showing the received sensor data wirelessly using Blynk app and (d) sensor output response graph using finger imparting.