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Strain Induced Rolled Thin Films for Lightweight Tubular Thermoelectric Generators

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Abstract

Thermoelectric generators (TEGs) are interesting energy harvesters from otherwise wasted heat. Here we show, a polymer assisted generic process and its mechanics to obtain sputtered thermoelectric (TE) telluride materials based 3D tubular structures with unprecedented length (up to seamless 4 cm and further expandable). This length allows for large temperature differences between the hot and cold ends, a critical but untapped enabler for high power generation. Compared to flat slab, better area efficiency has observed for rolled tube and compared to a solid rod architecture, a rolled tube uses less material (thus making it lightweight and cost effective) and has competitive performance advantage due to lesser contact area. We also show tubular architecture thermopile based TEG is able to achieve a generated power up to 5 μ W (8 pairs of p- and n-type thermopiles) through temperature difference of 60 $^{\circ}$ C. Demonstrated process can play important role to transform 2D atomic crystal structure TE materials into 3D tubular thermopiles for effective TEG application which can maintain higher temperature difference by longer distance between hot and cold end.

Keywords: Thermoelectric generator, Tube, Polymer, Strain Engineering, Power factor.

1. Introduction

Thermoelectric (TE) materials are of great interest due to their ability to transform thermal gradient between hot and cold zones of a system directly into electricity for their potential applications e.g., solid-state cooling devices, infrared detectors, and power generators, as a clean source for energy harvesting.^[1] Along with their reliability due to the absence of moving parts, relatively long life and silent operation, integration of thin film based thermoelectric generators in micro-scale systems provide advantages such as scaling and portability for various applications.^[2] In order to provide design flexibility for various microsystems, high-throughput fabrication technology with unparalleled yield and uniformity are needed on a variety of substrates for flexible electronics.^[3] In that regard, Telluride (Te) based TE alloy material are known for its room temperature stability and ease in deposition, high electrical conductivity and low thermal conductivity. Thus, high quality TE devices can be attained by using Antimony Telluride (Sb_2Te_3) and Bismuth Telluride (Bi_2Te_3) in various forms, including single crystal, thin films and different nanostructures.^[1, 4] Many techniques has been used to fabricate these in thin film forms, such as physical vapor deposition (PVD), metal organic chemical vapor deposition (MOCVD), and molecular beam epitaxy (MBE).^[1] Compared to MOCVD or MBE, sputtering is preferred for deposition of these TE materials, due to its ease of process control with its high throughput and reliability, whose deposition thickness is limited to achieve longer thermopiles. For longer thermopiles, whereas in-plane configuration is easily available by lateral deposition or coating, same plane surface hurts the thermal gradient by in-plane heat conduction and thermal convection between two zones of varied temperatures. Therefore, achieving a vertically aligned out-of-plane thermopiles would eliminate such heat convection induced performance compromise. Another demerit of conventional TE devices is they have high density thus they are heavy, which somehow restricts there use in energy harvesting applications.^[5] Here we show, a polymer assisted strain induced structural

transformation of thin films of telluride based TE materials into an unprecedented 4 cm long seamless 3D lightweight tubular architecture. The strain induced self-rolled tube is attained through formation of the tensile/stressed layer on the desired thin film (to be rolled) and then is lifted off with the optimized chemistry and mechanism of dissolving sacrificial layer from desired substrate. Both the approaches, bottom-up as well as top-down, are explained for the formation of self-rolling semiconductor micro-tubes and nanotubes structure.^[6-14] Bottom-up aspect is for high quality strained layers with appropriate composition and thickness, whereas the top-down aspect expose sidewalls of the desired thin film for lateral etching of the sacrificial layer in order to release the strained thin layer along with the desired TE film from the subsequent substrate.^[7-14] Later approach determines the precise tube orientation, dimensions and the number of rotations as per application prospects of TE applications. As a result, self-rolling tubular micro- and nanostructures can be assembled in large arrays with uniform diameters and exceptional spatial placement controllability. In the past, although researchers have focused on sensors, battery technology, and cellular vehicles applications using such methodology, this is the first time such concept has been used for TE applications.^[4-14] Additionally, the design aspects of tubular architecture, such as length and diameter are studied for potential impact on efficiency of thin film TE based devices. This tubular architecture based thermopiles was also demonstrated to be able to generate power for potential thermoelectric generator (TEG).

2. Results and discussion

Various process conditions (with TE film and stressor layer) were tested to optimize the process condition for the TE film based self-rolled tube architecture. To understand and carry the controlled self-rolling behavior of TE films, we optimized the process parameters in the following manner. As long as we have sputtered TE film on top of the SiO₂/Si substrate,

following key steps were observed: (i) as-deposited TE film has the tendency to roll up due to its inherent strained characteristic but in random and uncontrolled manner, and due to highly brittle nature it's tough to process the floating film, also it is a challenging aspect of this self-rolled architecture to study the stand alone/ free-standing architecture due to its low thickness and highly fragile nature; (ii) spin-coated stressor (SU-8 polymer) layer provides the controlled strained based rolling as well as freestanding bilayer architecture; (iii) heat treatment (baking) of entire structure (SU-8/thin film/SiO₂/Si) helps to maintain the strong adhesion and bonding between top two layers (SU8 over TE film), otherwise stressor layer comes off individually while rinsing the film in HF acid solution; (iv) stressor layer thickness (preferably few microns) and baking temperature (~ 95 °C) has been optimized as process control parameters. Thickness of the stressor layer found to be the most critical which determines the diameter of the strain induced TE self-rolled tube. It is observed that the most critical enablers to make the self-rolled TE films are thickness of stressed SU-8 layer, post growth heat treatment after the growth of sacrificial layer (~ 95 °C) and etchant of sacrificial layer (Conc. HF ~ 49 %). An interesting phenomena was also observed during the selective etching of sacrificial layer, after immersing the desired sample into HF solution for couple of minutes, samples were needed to gently pour into the aqueous medium for another few minutes (depending on the thickness of the stressor layer). As soon as the desired sample was taken out of the aqueous media, the bilayer (TE film with SU-8 layer) appeared to be self-rolled from the etched direction. As illustrated in **Figure 1**, for a TE thin film grown on sacrificial SiO₂ layer on Si substrate, the top strained SU-8 layer curves up along with desired TE film as soon as the sacrificial layer starts etching. TE layer starts rolling upon releasing from the substrate by selectively removing the SU-8 sacrificial layer, driven by the internal strain. This lift-off process is also explored to avoid the cost of Si wafers by reusing it as substrate. Overall yield was significantly higher (80% or above) for achieved rolled tube.

Crystal structure and the orientation of atomic crystals are determined using XRD measurement analysis for the 2θ range of $20 - 55^\circ$. The XRD patterns obtained for the as-prepared film confirm the existence of the rhombohedral Sb_2Te_3 phase (JCPDS 15-0874). Further, microscopic studies were carried out to observe the surface morphology of the sputter deposited TE film on SiO_2 . Figure 2 shows Scanning Electron Microscope (SEM) images of a typical sample of sputtered Sb_2Te_3 thin film deposited on SiO_2 at room temperature (a) in-plane top-view (b) cross-sectional view side view of the self-rolled film, and (c) typical XRD pattern of sputtered Sb_2Te_3 thin film deposited on SiO_2/Si , where peaks apart from Sb_2Te_3 structure shows reflection with sharp peak at 32.7° corresponds to Si (JCPDS 39-0973). Uniformly distributed grains with size of around 100 nm were observed through SEM technique.

Figure 3 shows images of self-rolled sputtered Sb_2Te_3 thin film (a) optical image of self-rolled tube architecture (b) cross-sectional optical view of self-rolled tube (c) side optical view of self-rolled tube, and (d) cross-sectional SEM view of self-rolled tube (shows different layer of Sb_2Te_3 film with SU-8 polymer stressor layer. Surface morphology shows that sputter deposited films at room temperature has uniform distribution of TE materials on SiO_2 substrate. Figure 3 (e) shows effect of SU-8 stressor layer thickness on the variation of the self-rolled Sb_2Te_3 tube diameter. The mechanics of stress induced thin film layer to 3D transformation shows the good fit of experimental data with predicted theoretical model which will be discussed in detail in next section.

Thermal transport properties has been carried out to understand the TE characteristic behavior of tubular thin films. For the fabrication of flexible TE devices, various polymer substrate can be used since it is flexible, have low thermal conductivity and remains stable over a specific range of temperatures. As as-deposited free standing TE thin films themselves have brittle structural behavior, process conditions are optimized for polymer layer which supports the sputtered thin film to study the characteristic for thermal transport analysis.

The efficiency of TE devices is associated with the dimensionless figure of merit (ZT) of TE materials, defined as; $ZT = (\alpha^2 \sigma / \kappa) * T$, where α , σ , κ and T are the Seebeck coefficient, electrical conductivity, thermal conductivity and absolute temperature, respectively.^[15] The crucial properties to understand thermal transport behavior i.e., Seebeck coefficient, electrical conductivity and thermal conductivity, of a typical Sb_2Te_3 based rolled tube architecture was found to be approximately $135 \mu\text{VK}^{-1}$, $4.4 \times 10^3 \text{ Sm}^{-1}$ and $10 \text{ WK}^{-1}\text{m}^{-1}$, respectively at room temperature. The respective power factor ($\alpha^2 \sigma$) is obtained in the range of $(7 - 9) \times 10^{-4} \text{ Wm}^{-1}\text{K}^{-2}$ over the temperature range of 270 K – 330 K, which is comparable to the reported literature values.^[16-18] The achieved TE parameters could be further improved by introducing different materials, compositional, alloy combination, growth conditions and dimensional approach. The detailed TE properties measurement is shown in Figure 4, which shows thermal transport properties variation of Sb_2Te_3 film based tube architecture as a function of temperature in Kelvin (a) Schematic of self-rolled tube sample with four contact probes (two for heater shoe and heat sink, rest two are of hot and cold thermocouples, respectively) for PPMS studies where inset shows the optical image of the rolled tube with four contacts, (b) thermal conductivity and electrical conductivity, (c) Seebeck coefficient, and (d) power factor respectively. Similarly, for a typical Bi_2Te_3 based rolled tube architecture, the thermal transport parameters observed as, $\alpha \sim -41.5 \mu\text{VK}^{-1}$, $\sigma \sim 1.3 \times 10^4 \text{ Sm}^{-1}$ and $\kappa \sim 19.1 \text{ WK}^{-1}\text{m}^{-1}$, respectively at room temperature.

3. Mechanics of stress induced 2D to 3D transformation

To manipulate the electrons for semiconductor based heterojunctions, strain induced self-rolled semiconductor tube could be one of the challenging architecture. As we understood, these self-rolled tubes are formed spontaneously as a result of energy minimization when a strained planar structure deforms into curved surfaces by strain relaxation.^[19] It can be inferred from the strain-induced thin film to 3D transformation for self-rolling mechanism illustrated in Figure. 1, that

rolling direction of the bilayer will be entirely dependent on the strained layer (compressive or tensile) deposited on the planar structure. In our work, the stressor layer is deposited on the top of the TE materials, such that, uncontrolled and/or inverted-rolling of the rectangular stripe patterns could be avoided. Special care has to be taken that the SiO₂ sacrificial layer can be easily etched in highly concentrated HF-based etchant thereby releasing the bilayer from the substrate. After etching the sacrificial oxide layer, the compressed layer stretches and develops an elastic force F_1 . On the other hand, the stretched layer compresses and develops an elastic force F_2 . The directions of forces F_1 and F_2 are opposite and create a non-zero moment (bending moment) of force M .^[7, 10] This bending moment is what transforms thin film patterns into 3D curved structures. The diameter of the self-rolled tube thus obtained depends on the thicknesses of the materials involved, their Young's modulus and the difference in stress.^[7, 10] The diameter of curvature is obtained by balancing the bending moment on the surface with the flexural rigidity as shown by Timoshenko in 1925.^[20] The equation for the diameter of the tube can be obtained as:^[21-23]

$$D = \frac{\left(\frac{E_1}{E_2}\right)t_1^4 + 4t_1^3t_2 + 6t_1^2t_2^2 + 4t_1t_2^3 + \left(\frac{E_2}{E_1}\right)t_2^4}{6\Delta\epsilon t_1t_2(t_1+t_2)} \dots\dots\dots [1]$$

Where, E and t are the Young's modulus and the thickness of the films and subscripts 1 and 2 denote the SU-8 and TE film. $\Delta\epsilon$ denotes the relative stress between the two films. For experimental verification of this model, the thickness of the TE was kept constant (Equation 1). It was observed that the diameter of the tubes varies with the thickness of the SU-8 stressor layer. The variation in diameter of the tube with thickness of SU-8 layer is as shown in Figure 3 (e). The points denote the experimental values while the solid line shows the theoretical prediction from above Equation (1). It can be observed that the experimental values closely match the theoretical prediction for a relative strain of 0.27 %. The strain mismatch was used as a fitting parameter, value of 0.27% was obtained by optimizing the least square error

between the calculated and experimental results. The growth of the specific strained layers with defined thickness and mismatch strain determines the self-rolled tube diameter. The length of the self-rolled tube could easily be designed as per the requirement. We have dealt the self-rolled tube architecture of the length up to 4 cm. The lift-off process is carried out through etching of sacrificial layer, with specified length and breadth of rectangular substrates, further helps in positioning and alignment control of these tubes.

The rolled TE architecture validation and analysis of our approach was performed through FEA simulation (shown in Figure 5). The simulation compares the different TE responses due to architectural difference. The simulation is for an open circuit, where applied voltage is the similar but resistances are different depending on the internal resistance which is a function of the cross sectional area (contact area in our case). Figures 5 (a, b) show the setup temperature gradient and the resulting induced potential distribution for a solid cylinder (rod), respectively. Similarly, Figures 5 (c, d) show the temperature gradient and induced potential for a hollow cylinder (tube). The temperature difference between the bottom and top of the structures is 280 degrees (an arbitrary value to induce significant voltage change). The system has adiabatic boundary conditions (no energy exchange with surroundings) and is composed of top and bottom alumina slabs at specific temperatures, copper contacts for current flow and the TE structures. Each TEG is composed of p-type and n-type TE material with the same Seebeck coefficients as function of temperature but with negative values for the n-type for simplicity. The FEA results show that the tube (hollow cylinder) architecture does not exhibit any degradation in the generated voltage compared to the rod (solid cylinder) architecture (for more details supplementary information is provided with Figure S1). Also, simulations are compared for flat version of the tube (as if a rolled tube has been cut along its length) (tube kept horizontally) with vertical tube on alumina for which TE response is ~ 4% lower in case of flat version than that of vertical arrangement (figure not shown).

It is observed that the thermal conductivity decreases as the shell thickness decreases, which is consistent with the thermal conductivity results for both thin film and simple nanowires due to the nature of ballistic phonon transport.^[24] The increased surface or interface area shows decrease in thermal conductivity of such nanostructures. Slight decrease of power factor is reported when the core-shell structure is introduced, associated with remarkable reduction of phonon thermal conductance.^[25] Compared to flat slab, better area efficiency has observed for rolled tube and compared to a solid rod architecture, a rolled tube uses less material which may make it lightweight as well as cost effective and has competitive performance advantage due to lesser contact area (for more simulation details supplementary information is provided with Figure S2). Including the architectural advantages, our methodology shows obvious less consumption of TE materials due its thin film nature compared to the bulk traditional thermopiles thus applicable in lightweight TE applications.

4. Output performance of tube based TEG

We have also demonstrated the TEG's based on the rolled films carrying p- and n-type tubes of Sb_2Te_3 and Bi_2Te_3 TE materials, respectively, all connected to each other in series with Au-coated Cu wires. Two micro-probes connected to a Keithley multimeter were landed on top of the two end contacts of the TEG. Figure 6 (a) shows schematic of typical tube-TEG power generation with two pair of p- and n-type legs. Figure 6 (b) shows the digital image of tube architecture based TEG with typical two pairs of p- and n-type legs whereas Figure 6 (c) reflects the power output for different combinations of p-type and n-type legs (2, 5 and 8 pairs respectively) with change in temperature (carried out with TE temperature controller). Our analysis shows that the output power density depends on the number of TE legs, their dimension and properties. For tubular architecture, lightweight tube-TEG with various TE legs is able to achieve a generated power up to 5 μW (8p + 8n) through maximum temperature

difference of 60 °C. The power increases with increase in temperature gradient up to the saturation point around ~ 85 °C. The change in the power output for 2 to 5 pairs are slightly higher compared to 5 to 8 pairs respectively, possibly due to increased contact resistance between the legs. This architecture satisfies the requirement for many micro-scale TE devices and is particularly important as it has been achieved without any special surface processing or post-deposition annealing.

5. Conclusion

We have demonstrated a generic fabrication process and associated mechanics for polymer assisted strain induced transformation of a 2D thin TE film into non-planar seamless 4 cm long 3D tubular architecture TE tube. The sputtered TE film of Sb_2Te_3 material has been deposited on sacrificial layer of thermally grown SiO_2 on Si substrate, at room temperature, which further coated with SU-8 stressor layer with optimized condition to occur self-rolling of bilayer with the etching of sacrificial oxide layer. We observed that the most critical parameters to make the self-rolled TE films are type of stressor layer as well as sacrificial layer, thickness of stressor layer, post growth heat treatment after the deposition of sacrificial layer, type of etchant for sacrificial layer and the optimization of time-interval for etching. The effect of stressor layer thickness is elaborated in terms of self-rolled tube diameters. The formation process of strain-induced self-rolled TE tubes are demonstrated in detail. We have studied the thermal transport behavior of rolled TE tube (single layer rolling), *i.e.*, Seebeck coefficient, electrical conductivity and thermal conductivity, of a typical Sb_2Te_3 based rolled tube was found to be approximately $135 \mu\text{VK}^{-1}$, $4.4 \times 10^3 \text{ Sm}^{-1}$ and $10 \text{ WK}^{-1}\text{m}^{-1}$, respectively at room temperature. The respective power factor is obtained in the range of $(7 - 9) \times 10^{-4} \text{ Wm}^{-1}\text{K}^{-2}$ over the temperature range of 270 K – 330 K, which is comparable to the reported literature values. Mechanics of stress induced 2D layer to 3D transformation shows the good fit of experimental

data with predicted theoretical model. The finite element analysis show that the tube architecture does not exhibit any degradation in the generated voltage compared to the rod architecture. Electrical studies show that for tubular architecture, lightweight tube based TEG, with various TE legs is able to achieve a generated power up to 5 μW (8 pairs of p- and n-type) through maximum temperature difference of 60 $^{\circ}\text{C}$. The self-rolled TE films could be made into arrays to fabricate efficient TE generators for energy harvesting applications. This process could be further improvised for the various architectural designs to relevant applications especially with 2D atomic crystal structure of efficient TE materials.

6. Experimental Section

In this work, a p-type lightly doped silicon wafer (4") has been used as a base substrate to perform the desired self-rolled TE tube device structure. Thin layer of silicon dioxide (SiO_2) was thermally grown (wet oxidation) at the temperature of 1000 $^{\circ}\text{C}$ with the thickness of 300 nm. We report the fabrication parameters for thickness $\sim 2.5 \mu\text{m}$ of Sb_2Te_3 and Bi_2Te_3 thin films (targets supplied by Angstrom Sciences) deposited on thermally grown SiO_2 layer at room temperature using RF magnetron sputtering system (NEXDEP 120). Optimal deposition conditions were found to be at 5 mTorr sputtering pressure, 10 sccm of Argon gas flow, sputtering power of 32 W, 7 cm distance between the sample and target, and 20 rpm substrate rotation respectively.

To create a stressor layer, we have used widely studied epoxy based negative photoresist polymeric material: SU-8TM. Compared to other commonly used polymeric materials such as Polydimethylsiloxane (PDMS), Poly(methyl methacrylate) (PMMA), polyimide, etc., it can be processed very easily, it has small thermal conductivity $\sim 0.2 \text{ W m}^{-1} \text{ K}^{-1}$, comparable modulus of elasticity with both Bi_2Te_3 and Sb_2Te_3 TE materials and its stress can be controlled easily. In addition, it is mechanically/chemically stable, chemically resistive

thus not dissolvable in acid, and it has relatively high thermal stability $>200\text{ }^{\circ}\text{C}$ and finally it is highly transparent. We used spin coating technique to deposit the SU-8 stressor layer on top of sputtered TE film. Standard coating process parameters (spin speed and baking time) has been followed to coat various thicknesses of SU-8 layers. Post growth heat treatment (baking) of coated SU-8 layer is performed on routine hot plate for stress formation at the temperature of $\sim 95\text{ }^{\circ}\text{C}$. To release the bilayer (TE film with stressor layer) in controlled rolling behavior, immersion of prepared sample into wet hydrofluoric (HF) etchant (concentration $\sim 49\%$) followed by aqueous medium (H_2O) are carried out for the removal of the sacrificial oxide layer. The microstructural characteristics of the thin films were investigated using Scanning Electron Microscopy (SEM) and X-ray Diffraction (XRD) techniques. The thermal transport properties, with respect to Seebeck coefficient, electrical resistivity and thermal conductivity, of these thin film based rolled tube were carried out with physical property measurement system (PPMS) equipment at specific range of temperatures. Gold coated copper wires were connected to the sample with epoxy paste for four necessary contacts to perform the TE property measurement. These thermal transport bars (conducting wires) are directly purchased from PPMS tool supplier (Quantum Design). Purpose of Au coated Cu - wire is for achieving good thermal as well as electrical contact with the sample. The rolled TE architecture validation and analysis of our approach was performed through Finite Element Analysis (FEA) simulation [COMSOL Multiphysics software package]. A 6 $\frac{1}{2}$ -digit Agilent multimeter was used to measure the voltage and current thus power while the gradient of temperature was set with a TE temperature controller.

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Author contributions

MH conceptualized and directed the study. DS carried out the experiment. AK assisted in visualization. AH worked on the mechanics. MG carried out the FEA modeling.

Supplementary information

All the supplementary information is available at
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Competing financial interests

We declare no competing financial interest.

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Figures

Figure 1. Process schematic of simultaneous operation of stress induced structure transformation and lift-off process enabling the formation of 3D tubular thermoelectric materials from a 2D thin film.

Figure 2. SEM images of a typical sample of sputtered Sb_2Te_3 thin film deposited on SiO_2 at RT (a) in-plane top-view and (b) cross-sectional view, and (c) XRD pattern which shows various peaks of Sb_2Te_3 films along with Si with different 2θ positions.

Figure 3. Self-rolled sputtered Sb_2Te_3 thin film based (a) optical image of 4 cm long self-rolled tube architecture; (b) cross-sectional optical view of the tube; (c) side optical view of the tube, (d) cross-sectional SEM view of the tube (shows different layer of Sb_2Te_3 film with stressor SU-8 polymer stressor layer), and (e) variation of the self-rolled Sb_2Te_3 tube diameter with the change in SU-8 stressor layer thickness (inset show tubes with two different diameters).

Figure. 4. Thermal transport properties variation of Sb_2Te_3 film based tube architecture as a function of temperature in Kelvin (a) Schematic of self-rolled tube sample with four contact probes (two for heater shoe and heat sink, rest two are of hot and cold thermocouples, respectively) for PPMS analysis where inset shows the optical image of the rolled tube with four contacts, (b) thermal conductivity and electrical conductivity, (c) Seebeck coefficient, and (d) power factor, respectively.

Figure 5. FEA simulation for two different thermoelectric material based architectures. (a, b) The setup temperature gradient and the resulting induced potential distribution for a solid cylinder (rod), respectively. Similarly, (c, d) show the temperature gradient and induced potential for a hollow cylinder (tube).

Figure 6. (a) Schematic of typical tube-TEG power generation with two pair of p- and n-type legs. (b) Digital image of tube architecture based TEG with typical two pairs of p- and n-type legs. (c) Power output for different combinations of p-type and n-type legs (2, 5 and 8 pairs respectively) with change in temperature.

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