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Paper-Based Origami Flexible and Foldable Thermoelectric Nanogenerator

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Abstract

Paper has been an essential material in our daily life since ancient times. Its affordability, accessibility, adaptability, workability and its easiness of usage makes it an attractive structural material to develop many kind of technologies such as flexible electronics, energy storage and harvesting devices. Additionally, the scientific community has increased its interest on waste heat as an environmentally friendly energy source to support the increasing energy demand. Therefore, in this paper we described two affordable and flexible thermoelectric nanogenerators (TEGs) developed on paper substrates by the usage of simple micromachining and microfabrication techniques. Moreover, they exhibit mechanical stability and adaptability (through folding and cutting techniques) for a diverse set of scenarios where vertical or horizontal schemes can be conveniently used depending on the final application. The first TEG device, implemented on standard paper, generated a power of

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0.5 nW (ΔT = 50 K). By changing the substrate to a tearless and extra-smooth polyester paper, the TEG performance was optimized achieving less internal resistance and a greater power of ~80 nW (ΔT = 75 K), at the cost of more rigidity in the substrate. This power represented over three times higher power production than the standard paper–based TEG with same dimensions, number of thermoelectric pairs and temperature difference. Another interesting aspect of paper based TEG is due to its foldability, one can control the temperature difference by unfolding (larger separation between hot and cold ends) and folding (smaller separation). Finally, one of the underlying objectives of this work is to spread the availability of essential technologies to the broad population by inclusion of everyday materials and simple processes.

**Graphical Abstract**

**Keywords:** Flexible, foldable, thermoelectric nanogenerator, paper, waste heat.
1. Introduction

From ancient times, paper has been an important platform and resource not only for communications and artistry, but it also has become an indispensable utility for innumerable tasks in our daily life. More recently, there has been an increasing interest to employ it as substrate to build low-cost and flexible electronics, all thanks to its affordability, accessibility, adaptability, workability and its easiness of usage [1–11]. Printing techniques have been previously used for this purpose, in which different materials can be successfully integrated with the paper substrate. Such materials can exhibit a wide range of electric properties, from conductive to semiconductive, and are used to fulfill diverse functions [12–14]. Nevertheless, the performance of these kind of printed electronics on paper substrates still have a lot of room for improvement that can lead to a more successful technology translation.

Moreover, there has been diverse efforts to combine distinctive materials with the paper itself for it to acquire conductive or semiconductive characteristics and become an active electronic material [15–18]. For example, researchers have made use of this approach to implement and demonstrate diverse applications such displays, sensors, radio frequency identification tags (RFID), energy harvesting and storage devices [19–32]. Particularly, paper-based energy harvesting devices have been developed to make use of mechanical or thermal energy, electrostatics, the triboelectric effect and others, mainly by functionalizing the paper with integrated nanomaterials or other metallic/polymeric composites [26, 33–35]. In this work, we are especially interested in the re-use and harvest of thermal energy because it is considered as one of the most important and environmentally benign sources of wasted energy that could be used to aid green energy production [36–39]. Current efforts towards the

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optimization of systems that can take advantage of waste heat have been directed to improve and develop novel materials and composites [38, 40]. Nevertheless, there is still a large gap in terms of finding an affordable and cost-effective solution to achieve commercial success [41]. Exceptionally, applications in wearable electronics (i.e. an innovative, rising technological paradigm in smart living) can be hugely benefited from harvesting systems that use the human body’s heat to generate electricity and to power up sensors integrated into a wearable system for both monitoring and actuation [42–45]. An important additional characteristic appropriate for wearable technologies is the ability of bending and stretching to conform to the shape of a flexible object. Therefore, the final implementation of an energy harvester depends not only on performance but also on mechanical conformability, flexibility, and affordability. With this set of principles in mind, a balanced solution must be established and an adequate set of materials must be chosen.

Polymers/organics, as either active or structural materials, are valid and important alternatives to paper and several examples of flexible, polymeric/organic-based thermoelectric generators (TEGs) have been demonstrated [46–49]. The use of polymer-based substrates to support thermoelectric (TE) materials is of special interest since highly advantageous inorganic materials can be integrated, including those with superior thermal and electrical properties [50]. An important advantage of polymer-based substrates, such polyimide (PI) [51] and SU8 [52], is their capability to form out of plane structures [53,54] and be used with more advanced photolithography-based fabrication techniques, which can offer greater resolutions, hence allowing the creation of more complex and compact implementations [51–54]. On the other hand, paper is a remarkable structural material which can be used to fabricate simple, affordable and flexible TEGs [55]. Moreover, very interesting and useful origami and kirigami techniques can be employed with paper substrates to implement devices with enhanced functionality and for diverse applications [56–58].
Nevertheless, approaches to use paper as substrate and structural material, supporting active and high-performance inorganic TE materials, has not been developed yet. This paper-based approach is interesting, given the low thermal and electrical conductivity of the paper and its advantageous mechanical properties. In this work we have used simple and scalable micromachining and microfabrication techniques to integrate inorganic TE materials, exhibiting high figures of merit (ZT), with two kinds of paper for a diverse set of scenarios where vertical or horizontal schemes can be conveniently used depending on the final application. In the first example, we show a foldable TEG with adjustable height for area efficient implementations, which produced a power of 0.5 nW at a temperature difference of 50 K. The second implementation consists of a tearless, extra-smooth paper-based TEG with improved resistance and power generation, and the possibility of vertical and horizontal placement. The power achieved with this approach was ~80 nW at a temperature difference of 75 K, which represented over two orders of magnitude higher power production compared with the first implementation (0.5 nW @ \( \Delta T = 50 \) K), and over three times greater power production compared to a standard paper–based TEG with the same dimensions, number of TE-pairs and temperature difference (25 nW @ \( \Delta T = 75 \) K).

2. Material and Methods

We have selected two different kinds of paper, a standard paper and a stiffer, waterproof, tear-resistant, heat stable and extra-smooth paper (Permanent or Polyester Paper) as substrates. Bismuth telluride (\( \text{Bi}_2\text{Te}_3 \)) and antimony telluride (\( \text{Sb}_2\text{Te}_3 \)) are well-known TE materials with high figures of merit that have been often used to implement efficient TEGs for an extensive range of applications. Their popularity is in part due to their ability to be integrated on top of several kinds of substrates by diverse deposition methods [50, 59–61]. Consequently, we have used them as the active TE materials of our TEG implementations.
In the case of the first implementation, a standard paper has been used as a substrate to host four thermopiles \((\text{Bi}_2\text{Te}_3\text{-Sb}_2\text{Te}_3 \text{ pairs})\) as shown in Figure 1. This design features a simple and rapid fabrication and implementation with adjustable TEG height by paper folding, thus achieving a higher temperature difference. In the first place, strips of TE materials (2 mm wide, ~3.5 cm long) were sequentially deposited by PVD magnetron sputtering (Angstrom Engineering, NEXDEP CS-05 loaded with Bi\(_2\)Te\(_3\) and Sb\(_2\)Te\(_3\) targets; Deposition parameters: 10 sccm of Ar flow, 5 mTorr pressure, 20 rpm substrate rotation and 30 W power at 20 °C) to a final thickness of ~750 nm with the assistance of polyimide hard masks. Figures 1a and 1b show scanning electron microscopy (SEM) images of one Bi\(_2\)Te\(_3\) strip, showing full coverage above and in-between the paper fibers (black spots in the images are due to charging effects rather than non-coverag or delamination of the TE material). Finally, gold strips were deposited to connect the TE pairs and to act as contact pads (20 nm/300 nm of sputtered Ti/Au). Hence, the scheme consists of a foldable piece of paper (4 cm × 2 cm), on top of which the TE pairs were formed. The paper can be then folded or stacked, which allows area saving and adjusting the separation between hot and cold sides of the nanogenerator, thus increasing the temperature gradient between junctions at the device level.

In an effort to optimize power generation, the second implementation consisted of adding more pairs and reducing the internal resistance considerably. The latter was possible due to the smoother surface of the polyester paper and the formation of shorter and thicker TE legs/strips (1 mm wide, 8 mm long, ~2 μm thick). Two set of devices were fabricated, one with 10 TE pairs and one with 20 pairs as shown in Figures 2a and 2b (Total area in both cases was 4 cm × 1 cm). The fabrication process was very similar to the first version with the exception that the shadow masks were defined on a vinyl paper sticker material using a micromachining CO\(_2\) engraving laser (Universal PLS6.7S), emitting a laser wavelength of 10.6 μm and delivering a maximum power of 75 W [62, 63]. The adhesive material was
preferred because higher resolution features can be more defined during the TE depositions, as the shadow mask is in direct and fixed contact with the substrate. On the other hand, the laser cutter allowed higher dimensions and spacing precision and better alignment accuracy.

The sputtering deposition of the TE materials was sequentially performed as described before and gold paths were skipped for simplicity, so connections were done directly on the TE materials. Figures 2c and 2d show the SEM images of the Bi₂Te₃ layer on top of the polyester paper. The film has a more uniform and smoother surface with the absence of fibers but leaves microscopic lumps inherent to the paper itself.

TEGs were also simultaneously fabricated with the same design on standard paper so we could compare their performance with the ones that were fabricated on the polyester paper.

A hot plate with a digital thermometer and a 6 ½-digit Agilent Multimeter were used for the electrical characterization of all the devices.

Finally, mechanical bending and cycling tests were also conducted and briefly discussed. A more detailed description can be found in the supplementary material.

3. Theory

The typical thermoelectric material figure of merit (ZT) is given by the following expression [39]:

\[
ZT = \frac{PF \cdot T}{\kappa} = \frac{S^2 \sigma T}{\kappa}
\]

where \((PF = S^2 \sigma)\) is the power factor, \(S\) is the Seebeck coefficient, \(\sigma\) is the electric conductivity, \(\kappa\) is the thermal conductivity and \(T\) is the temperature. Therefore, a good-performing thermoelectric material should accomplish three different objectives: (i) achieve a low thermal conductivity to maintain a larger temperature difference, (ii) reach a higher Seebeck coefficient to achieve higher Seebeck voltages and (iii) attain a higher electrical conductivity to get a larger short-circuit current. Altogether, this will lead to higher efficiency.
and power generation. The challenge remains in achieving all the goals at the same time as thermal and electrical conductivities are usually closely related. In nature, we can find both organic and inorganic material that exhibit good thermoelectric properties, but at this point inorganic materials still dominate with the highest ZT values (>1) [39, 49]. Among the inorganic materials commonly used in thermoelectric applications, Bi$_2$Te$_3$ and Sb$_2$Te$_3$ are two of the best known and given their ease of deposition through sputtering, we have selected them for our demonstrations. We have measured the Seebeck coefficients of both Bi$_2$Te$_3$ and Sb$_2$Te$_3$ thin films using a separate setup as described elsewhere [60]. The values are -37 $\mu$V/K for Bi$_2$Te$_3$ and 240 $\mu$V/K for Sb$_2$Te$_3$ at 314 K.

On the other hand, there have been several approaches to engineer new materials that achieve higher ZT values. The use of nanomaterials, nanocomposites or 1D/2D materials is of special interest since low-dimensional systems present several interesting effects to achieve higher ZTs [64–68]. First of all, the thermal conductivity can be decreased by increasing the boundary scattering of phonons. Additionally, the density of states is enhanced, increasing the Seebeck coefficient. Finally, an improved quantum confinement also helps improve the power factor. Our objective, though, is not to demonstrate the highest performance but rather to set the precedent that low-cost materials can be combined with simple techniques and innovative designs to develop affordable but efficient harvesting devices for waste heat, which can be implemented rapidly while simultaneously exhibiting flexibility and mechanical adaptability to different scenarios and applications.

4. Results

Figure 3 summarizes the main results of the paper-based TEGs developed in this work. Figure 3 shows the polarization curve and current-voltage (IV) curve of the standard paper-based TEG respectively, with a maximum power registered of about 0.5 nW at a difference in
temperature of 50 K. The internal resistance derived from the I-V curve was 1.25 MΩ, a significant value that may be linked with high porosity and surface topography of the standard paper and that helps to elucidate the low output current from the TEG.

In this first implementation, it is worth noting that only four TE pairs were used, so increasing the number of pairs would increase the power proportionally. With this in mind, for the second implementation, we tested two standard paper-based TEGs with 10 and 20 TE pairs. As expected, the TEG with 20 pairs produced about twice as much voltage as the 10 TE pairs version. A voltage value of 96.5 mV open circuit voltage (OCV) was obtained from the 10-pairs TEG, whereas 190.7 mV OCV was obtained from the 20-pairs TEG. However, we also noticed that the internal resistance increased (from 172 kΩ with 10 pairs to 425 kΩ with 20 pairs) and therefore the output current was impacted. In consequence the final power production did not actually reach twice the original value as one would initially expect (The maximum power for the 10-pairs TEG was ~15 nW at a difference in temperature of ~80 K and for the 20-pairs TEG was ~24 nW at a difference in temperature of 75 K). This observation tells us that the number of pairs is not necessarily linearly proportional to the power generation, and it is just as important to reduce the final resistance by optimizing the resistivity of the films and improving the thermoelectric properties. Subsequently, the selection for a better-quality, smoother-surface polyester paper had significant repercussions, which included less resistive deposited TE films and therefore a substantial improvement in power generation. Figures 4a and 4b show the polarization curve and IV curve of a 20-TE-pairs on polyester paper TEG that reached over 80 nW of power at 75 K, a power over three times higher than the one produced with the standard paper at the same temperature difference.
The achieved power represents 53.3 pW/K per TE-pair, a competitive value especially given the use of paper, a rough substrate, the simplified fabrication methods and the capability of efficiently folding/staking the paper-based TEGs to aggressively increase the power density.

The obtained resistivity of the films was found to be 8.6 $\mu\Omega\cdot$m for Bi$_2$Te$_3$ on the polyester paper and 13.4 $\mu\Omega\cdot$m for Bi$_2$Te$_3$ on standard paper. For Sb$_2$Te$_3$ film resistivity, the values found were 952 $\mu\Omega\cdot$m for polyester paper and 1232 $\mu\Omega\cdot$m for standard paper. Improved resistivity of the thin films and thicker depositions produced an internal resistance of $\sim$130 k$\Omega$, a three times lower resistance than what was achieved with the standard paper (425 k$\Omega$ as mentioned above).

We have calculated the power factors and ZT values according to Eq. (1) at 314 K and assuming a thermal conductivity of 2 W/m·K for Bi$_2$Te$_3$ [69] and 2.5 W/m·K for Sb$_2$Te$_3$ [70]. For standard paper, the calculated power factors and ZT values were 102 $\mu$W/K$^2$·m and 0.016 for Bi$_2$Te$_3$ and 44.6 $\mu$W/K$^2$·m and 0.0056 for Sb$_2$Te$_3$. In contrast the calculated values of power factors and ZT on polyester paper were 160 $\mu$W/K$^2$·m and 0.025 for Bi$_2$Te$_3$ (~60% higher than on standard paper) and 60.5 $\mu$W/K$^2$·m and 0.0076 for Sb$_2$Te$_3$ (~35% higher than on standard paper).

It is important to notice that this kind of paper-based TEGs can easily be folded in diverse ways (e.g. accordion-like or fan-like, origami and kirigami techniques). This characteristic helps in the design of the TEGs either by drastically reducing the footprint and increasing the power density or by giving it an additional functionality to the device such as stretchability, foldability and other similar approaches commonly found in flexible electronics. More importantly, it opens up opportunity to control the distance between the hot and cold end by folding (shortening distance) and unfolding (expanding distance). In fact, a recent study shows how it is possible to achieve higher power generation thanks to the increase in temperature gradient by stretching a paper-based TEG fabricated with the same set of
materials and techniques. This is possible as long as the thermal conductivity of the substrate material is low enough, as is the case of the polyester paper [71].

Finally, we performed a compressive strain and cycling mechanical tests for each of the TE films deposited on both standard and polyester papers (detailed description and graphs are shown in the supplementary material). First, a compressive strain test (Fig. S1) showed some increment in electric resistance of the TE films on standard paper up to near 80% of applied strain after which there is a sharp degradation. On the polyester paper the Bi$_2$Te$_3$ film showed a stable and uncompromised behavior regardless of applied strain. However, the Sb$_2$Te$_3$ film displayed a sharp increase in electric resistance, abruptly jumping to near hundred times the initial value at 35% applied strain. Small cracks appeared on the film, which can explain the abrupt change in resistance. Therefore, even though the smoothness and firmness of the polyester paper were advantageous to reduce the resistivity of the deposited TE films, it made the films a little more prone to fracture due to the increased rigidity inherent with this kind of tearless paper. A cycling test was also conducted up to a maximum compressive strain of ~15% for 400/300 cycles on the standard/polyester papers for both TE films (Fig. S2). On standard paper, both films showed a small increment of around 2 times the original electric resistance after the 400 cycles. For the case of the polyester paper, a slightly higher increment in the electrical resistance was observed up to 10 and 15 times for the Sb$_2$Te$_3$ and Bi$_2$Te$_3$ respectively after 300 cycles. This increment also reflects the stiffer nature of the polyester paper.

5. Conclusions

In this work, we have used simple micromachining and microfabrication techniques to integrate inorganic materials with paper substrates to develop affordable and efficient thermoelectric nano-scale power generators. These devices can be implemented rapidly and,
at the same time, exhibit flexibility and mechanical adaptability for a diverse set of scenarios ranging from vertical or horizontal schemes that can be conveniently used for various applications. Two kinds of papers were used, standard and polyester-based, to demonstrate foldable, flexible and affordable TEGs. The first TEG implementation (on standard paper) presented a power of 0.5 nW at a temperature difference of 50 K, whereas an optimized second implementation (on a tearless, extra-smooth, polyester paper) achieved less internal resistance and a higher power generation (~80 nW) at a temperature difference of 75 K. This performance represents over three times higher power production compared with a standard paper–based TEG.

Mechanically, both TE films are stable on both paper substrates, showing some degradation in electric resistance after a compressive strain test or a cycling test, although due to the stiffer nature of polyester paper, the degradation can be more noticeable. Encapsulation and placement of the TE films in the neutral mechanical plane will help to reduce this effect.

One important advantage of this kind of paper-based TEGs is their ability to be folded in diverse ways (e.g. accordion-like or fan-like) to reduce the footprint of the device drastically and thereby increase the power density generation. Additionally, techniques such origami or kirigami can be used to offer additional functionalities to the devices including improved flexibility or even stretchability.
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References

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Vitae

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Ian Foulds completed his PhD in 2007 at Simon Fraser University, receiving the Governor General's Gold Medal. He then took up an NSERC Postdoctoral Fellowship at the University of Victoria; after which, he joined King Abdullah University of Science and Technology (KAUST) in 2009 as an assistant professor of electrical engineering. Ian is currently assistant professor at The University of British Columbia, School of Engineering, Okanagan Campus. His research interests are in the area of microsystems design and fabrication with special interest in the use of polymer materials, having worked extensively with different polymer resists and polymer specific patterning techniques.

Dr. Muhammad Mustafa Hussain (PhD, ECE, UT Austin, Dec 2005) is an Associate Professor in the Electrical Engineering Program of KAUST. Before that he was Program Manager of Emerging Technology Program in USA’s largest semiconductor research consortium, SEMATECH, Inc. Austin, Texas. His research interest is in development of integration strategy by rational design of materials, processes and devices for futuristic electronics and systems for smart living and a sustainable future. Recipient of 37 research awards, he is lead inventor for 50+ US patents (awarded and pending). He is a Fellow of American Physical Society, Institute of Physics and Institute of Nanotechnology (UK). He is also Editor of IEEE Transactions on Electron Devices and Editor-in-Chief of Applied Nanoscience.
Figures

**Figure 1.** **a)** Scanning electron microscopy (SEM) image of a strip of Bi$_2$Te$_3$ deposited on standard paper. **b)** Zoom-in SEM image of a). Standard paper–based foldable TEG with four thermoelectric pairs, **c)** top view, and **d)** side view. (In the middle, a schematic representation of the foldable paper-based TEG).
Figure 2. a) Polyester paper–based TEG with 20 thermoelectric pairs set horizontally, and b) with 10 thermoelectric pairs set vertically by cutting and folding. c) Scanning electron microscopy (SEM) image of a strip of Bi$_2$Te$_3$ deposited on polyester paper. d) Zoom-in SEM image of c). e) Vertically standing, standard paper–based TEG with 10 thermoelectric pairs and f) 20 thermoelectric pairs.
Figure 3. Plot of power (nW) and voltage (mV) versus current generated by the first implementation of a foldable, standard paper–based TEG.
Figure 4. a) Plot of power versus current and b) voltage versus current comparing the results from the second implementation of standard paper–based TEG and polyester paper–based TEG.
Research Highlights

- Paper based thermoelectric nanogenerators (TEGs) are demonstrated.
- Nano-scale power is generated which can be further enhanced by enhanced thermoelectric property of the material and design changes (including extended number of thermopiles and expanded distance between hot and cold end).
- Wrinkle free but more stiff polyester paper based TEG shows better performance than standard paper.
- Paper based TEGs with folding and unfolding can be used to control the distance between hot and cold side.