Sensors-on-paper: Fabrication of graphite thermal sensor arrays on cellulose paper for large area temperature mapping

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A R T I C L E   I N F O

Article history:
Received 23 August 2021
Received in revised form 29 October 2021
Accepted 7 December 2021

Keywords:
Thermal sensors
Thermocouples
Graphite sensors
Temperature mapping
Low-cost
Paper and pencil

A B S T R A C T

This paper reports on a fabrication method to obtain multiple thermal sensors by employing an array of graphite thermocouple patterns on commonly available Xerox paper. The graphite thermocouples are patterned using two different grade graphite pencils, which show a stable and reproducible thermal sensitivity. The fabricated paper devices with multiple thermocouple arrays are capable of producing temperature mapping of the desired area. Different thermal conditions were applied to test and confirm the working of these devices. The present work shows that simple graphite trace patterns can convert a piece of paper into a thermal mapping device.

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Specifications table

| Hardware name | Graphite thermal sensor array |
|---------------|-------------------------------|
| Subject area  | • Engineering and materials science |
|               | • General                      |
|               | • Measuring physical properties and in-lab sensors |
|               | • Field measurements and sensors |
|               | • Mechanical engineering and materials science |
| Hardware type | Thermal Imaging Camera |
| Closest commercial analog | CC BY 4.0 |
| Open source license | $ 0.34 |
| Cost of hardware | https://doi.org/10.17632/cg38djj8y5.2 |
| Source file repository | |
| OSHWA certification UID | |

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https://doi.org/10.1016/j.ohx.2021.e00252
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Temperature sensing is crucial in a wide variety of applications [1,2]. Different thermal sensors are being used in many industries including medical, agriculture, automotive, and so on [1,3–6]. Thermocouples are considered as a universal type of thermometers among other thermal sensors because of their simple configuration and passive nature [7–9]. A conventional thermocouple is made of two different metals/conductors, which are joined at one point to form a junction for thermal sensing [3,10]. As thermocouples are based on the Seebeck effect, dissimilar conductors of different thermopower ($S = \Delta V/\Delta T; \Delta V$ is the voltage generated from a conductor due to the temperature difference, $\Delta T$) are necessary to construct these devices [11,12]. The two conductors generate different voltages when the junction is heated; as a result, the thermocouple develops a net electrical voltage that can be used to measure temperature [13].

Fig. 1 shows a schematic structure of a traditional thermocouple with its working principle. If $V_{c_1}$ and $V_{c_2}$ are the voltages generated from conductor-1 (C1) and conductor-2 (C2), respectively, under a given temperature difference, $\Delta T$, then the net output voltage signal $(V)$ from the thermocouple is expressed as

$$V = V_{c_1} - V_{c_2}$$

(1)

where $V_{c_1} = S_1 \Delta T$ and $V_{c_2} = S_2 \Delta T$ ($S_1$ and $S_2$ are the thermopowers of conductors C1 and C2, respectively). Therefore, $V$ can also be expressed as: [14]

$$V = (S_1 - S_2) \Delta T = S_T \Delta T$$

(2)

The sensitivity of the thermocouple ($S_T = S_1 - S_2$) can be obtained from the ratio of net output voltage and temperature difference between the junction and surroundings.

Thin film-type thermocouples are attractive because of their lightweight structures and construction possibilities of flexible sensors [2,7]. Also, these film type thermocouples are suitable to construct planar sensor arrays which can be used to obtain the two-dimensional local temperature distribution of a surface or object [7]. Here, we have designed and built low-cost temperature mapping sensor arrays on commonly available Xerox paper using graphite pencils. The ability of different grade graphite pencils to show different thermopowers has been utilized to construct multiple sensor arrays of single conductor thermocouples in the form of completely graphite-based multiple sensor devices [11]. Typically, single-conductor thermocouples are fabricated from a single metal strip/layer, which requires two dissimilar width patterns of a metal of which at least one with sub-micron or nanoscale width in order to achieve different thermopowers from a single metal [10,14,15]. Such devices demand special fabrication techniques such as electron-beam lithography or photolithography in order to form sub-micron width metal patterns [7,16]. In addition, the multiple thermal sensor patterns are obtained by using special fabrication facilities, such as screen printing or thin film deposition techniques [1,15]. Here, a very simple method for constructing multiple sensor patterns has been demonstrated using commonly available graphite pencils and cellulose paper, and the presented method can be useful in the fabrication of low-cost thermal sensors. Also, the thermal sensitivities of these simple graphite sensors are comparable to single metal thermocouple sensor patterns [7,10].

2. Hardware description

Thin-film type thermocouple arrays are in general obtained from the deposition of two dissimilar conductor patterns on desired substrates [2,7,8]. Here, graphite-on-paper based thermal sensors are fabricated simply with hand drawing using two pencil grades (HB and 6B pencils). The HB and 6B grades were chosen to fabricate thermocouples as they produce a maximum thermal sensitivity in the resulting thermocouple sensors [11]. Graphite pencils (WH Smith) and commonly available office Xerox paper (80 g m$^{-2}$) were used to construct these multiple sensor arrays.

A single junction thermocouple was first fabricated in order to study its output characteristics and obtain the thermal sensitivity of the sensor. Two devices of 4-junction and 16-junction arrays were fabricated in order to demonstrate their successful functioning as local thermal distribution/mapping sensors. The 16-junction device consists of an array of 4 $\times$ 4 sensing junctions covering a thermal mapping area of 3 cm $\times$ 3 cm. Fig. 2 shows real pictures of the designed single-junction, 4-junction, and 16-junction sensor patterns on Xerox paper. An equivalent schematic of these sensors is depicted in Fig. 3 to show the distinctions between the HB and 6B pencil traces and the resulting junctions formed between them.
Fig. 2. Real pictures of the designed (a) single-junction, (b) 4-junction, and (c) 16-junction sensor patterns on Xerox paper. Every junction in the sensors is a combination of HB and 6B trace.

Fig. 3. Schematic illustrations of the designed (a) single-junction, (b) 4-junction, and (c) 16-junction sensor patterns. The yellow background represents the sensing area or temperature mapping area and a red dotted circle in (a) shows the junction of the thermocouple.

Fig. 4. Schematic illustrations of alternative designs of (a) 4-junction and (b) 16-junction multiple sensor arrays (the dotted red circles indicate junctions of the devices).
Fig. 5. Output voltage data of a single junction thermocouple with respect to temperature difference (ΔT).

Fig. 6. Relative thermopower variation of four graphite thermocouple junctions in a 2 × 2 sensor array (all data are normalized to the output of the J1 sensor).
Fig. 7. Output voltage data of three graphite sensor devices with respect to temperature difference ($\Delta T$).

Fig. 8. Temperature maps obtained from a graphite-based multiple sensor device ($4 \times 4$ array). (a & d) show two different heating configurations where the hot plate was heated to $\sim 60^\circ C$, (b & e) are the raw temperature data of the sensors, and (c & f) are the temperature maps obtained from a smooth interpolation method.
3. Design files summary

| Design file name | File type | Open source license | Location of the file |
|------------------|-----------|---------------------|----------------------|
| Fig. 1           | JPG       | CC BY 4.0           | Available with the article (https://doi.org/10.17632/cg38djj8y5.2) |
| Fig. 2           | JPG       | CC BY 4.0           | Available with the article (https://doi.org/10.17632/cg38djj8y5.2) |

Fig. 9. An illustration to show the measurement procedure for the multiple sensor array. V1, V2, V3, and V4 are the voltage outputs of the junction 1, 2, 3, and 4, respectively.

Fig. 10. Thermal sensitivity of a device as a function of bending cycles.
4. Bill of materials summary

| Designator          | Component                  | Number | Cost per unit - currency | Total cost - currency | Source of materials                                                                 | Material type |
|---------------------|----------------------------|--------|--------------------------|-----------------------|-------------------------------------------------------------------------------------|---------------|
| Graphite Pencils    | HB and 6B Pencils          | 2      | $ 0.17                   | $ 0.34                | https://www.whsmith.co.uk/products/whsmith-silver-hb-pencils-pack-of-12/5013872077181.html | Organic (Carbon) |

5. Build instructions

The construction of the graphite sensors involves the following simple steps:

As shown by Fig. 3 (a), the single junction sensor consists of a HB trace and a 6B trace overlapping at one end forming a junction between them. Pencil trace width of ~ 1 mm was maintained for both HB and 6B patterns and a length of 5–6 cm were used to construct all the sensor devices. A uniform and repeated pencil writing of about 20 cycles for each trace was performed to achieve good connectivity between the graphitic structures and reproducibility. A similar procedure was followed to fabricate multiple sensor arrays (Fig. 3 (b-c)) by keeping a space of 1 cm between the junctions, as a result, a 4-junction array covers an effective measuring area of 1 cm × 1 cm and a 16-junction array covers an area of 3 cm × 3 cm. The junction area of the sensor array was then covered with a polyimide tape, which acts as a protective coating for the junctions and improves the physical stability of the devices.

Alternative designs of multiple sensor arrays were also tested during the initial stage; the schematic structures of such designs are shown in Fig. 4. These devices also provide similar output information but the output leads are in all directions while the proposed designs (Fig. 3) have all the output leads on one side of the device making it convenient to read and collect the output data.

6. Operation instructions

Please refer “Validation and characterisation” section.
7. Validation and characterisation

The thermal sensitivity of a graphite thermocouple formed using HB and 6B pencil traces was obtained by measuring its output voltages at different heating temperatures of the junction. A simple lab-made measurement setup was used to achieve different heating temperatures [17]. Digital voltmeters (Model: HMC 8012 DMM) and standard temperature indicators were used for all the measurements, and measurements were repeated to confirm the reproducibility of the outputs. Fig. 5 shows the output characteristics of single junction graphite sensor. The output voltage data show a linear change with respect to temperature difference (∆T) applied to the device. A linear fitting of the data was carried out to obtain the thermal sensitivity of the sensor, which gave a slope value of 7.6 (±0.4) μV K⁻¹ with a coefficient of determination (R²) value of 0.995. This sensitivity has been used in the characterisation of multiple sensor array devices to represent their voltage outputs as temperature readings. These sensors can provide temperature information with a resolution better than 0.2 K with a microvoltmeter (as the micro-voltmeter can measure a minimum value of 1 μV, these graphite sensors with sensitivity of ~ 7 μV K⁻¹ can provide a temperature resolution of 1 μV / 7 μV K⁻¹ = 0.14 K, or approximately better than 0.2 K).

Fig. 6 shows the typical thermopower deviations of four individual junctions of the same multiple sensor device (2 x 2 array). The thermopower of these four graphite thermocouple junctions in a 2 x 2 array are all normalised to the reading of the first junction and all the sensors show very small deviation, which indicated good reproducibility of the sensor array. The reproducibility has been further confirmed by testing three devices; the output voltages shown in Fig. 7 indicate that these graphite based sensors can produce reproducible results.

The performance and reliability of the present graphite-on-paper multiple sensor arrays to obtain two-dimensional mapping of local temperatures has been demonstrated by fixing a 4 x 4-sensor array device on a hot metal plate. Two different heating configurations as shown in Fig. 8 (a & d) were used to test proper working of the sensor device. All the readings were measured when the corresponding hot plate temperature was nearly stable. The measurement procedure of these multiple sensor arrays is quite straightforward. The 4 x 4-array device consists of four columns each with four junctions. For every column, the first trace (for example, HB trace is the first trace in column 1) acts as a common lead for all the voltage outputs of the sensors in that column. Fig. 9 gives an illustration on the measurement procedure with highlighting the first column sensors. The same procedure was followed for all the columns to obtain 16 output voltage readings that are represented by Fig. 8. Also, these paper devices packaged inside the polyimide tape (Kapton tape) can be used multiple times without any degradation in the output properties. A device outputs have been studied after multiple bending cycles (Fig. 10) to confirm its reproducibility. The bending was performed along the length of the graphite traces with an approximate bending angle of 90°.

The sensitivity values of the present graphite based sensors are comparable to single metal based thermocouple sensors. For illustration, in Ni and Pd thin film based single metal thermocouples, sensitivities of about 5 μV K⁻¹ have been reported [7,10,18]. Higher thermal sensitivities of about 35–40 μV K⁻¹ have been reported in graphene based devices [1,15]. Similar improvements in the sensitivity can be expected from the present graphite based devices through the use of proper dopants [19]. In conclusion, the present study shows that these simple graphite trace patterns can convert a piece of paper into a thermal mapping device. The major elements of these sensors, the graphite and cellulose paper are abundant, low-cost, and non-toxic [20–25]. Therefore, extremely simple, cheap and disposable thermal sensors can be obtained with a simple pencil art. A more number of sensors can be accommodated on a specific area with standard thin film deposition techniques or screen-printing based methods to obtain temperature mapping of an area with high resolution. Further, the existing thermoelectric energy conversion devices/generators are usually manufactured from toxic and expensive semiconductors such as bismuth tellurides, lead tellurides, and their alloys [26–28]. Therefore, the current thermoelectric research is focused on finding low-cost and non-toxic thermoelectric conductors for the application [29–37]. With some improvements and modifications in the thermoelectric properties of graphite such as by organic doping or alloying with other semiconductors, they can also be utilized in the fabrication of thermoelectric devices [19,38–45].

CRediT authorship contribution statement

Rafiq Mulla: Writing – original draft, Conceptualization, Methodology, Visualization, Investigation, Validation. Charles W. Dunnill: Writing – review & editing, Supervision, Funding acquisition.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Acknowledgments

Authors are thankful to the Welsh Government (EU European Regional Development Fund) for funding the RICE (Reducing Industrial Carbon Emission) project (Grant Number: 81435).
Dr. Rafiq Mulla obtained his M.Sc. in Physics and got his Ph.D. from Karnatak University (2018, India) in Thermoelectrics. He is currently a Postdoctoral Researcher at Swansea University. His research has been focused on developing thermoelectric materials for Heat-to-Electricity and water splitting applications. Rafiq's research interests lie in the "lower-cost thermoelectrics", primarily developing new and economical synthesis methods, and cost-effective and less-toxic materials for the use of thermoelectric applications.

Dr. Charles W. Dunnill is a senior lecturer at Swansea University specializing in sustainable hydrogen generation technology. He was awarded his Ph.D. from Glasgow University in Nanomaterials before taking up a postdoctoral researcher post at University College London, where he was a Ramsay Fellow. Charlie advocates and researches the use of hydrogen as the perfect universal energy carrier to store, transport, and decouple renewable energy supplies from demand for energy, as well as photocatalytic water splitting as a method for solar energy harvesting.