

Fully Printed Sensors for In Situ Temperature, Heat Flow, and Thermal Conductivity Measurements in Flexible Devices

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ABSTRACT: Flexible temperature sensors allow temperature monitoring in wearable healthcare devices. A temperature sensor, which can be printed on flexible substrates, is designed and fabricated using a low-cost silver particle ink and a fast and scalable screen-printing process. A high temperature resolution of 10 m°C is reached. The versatility of this temperature sensor design is demonstrated for various applications, including in situ heat flux measurements, where a 2 mW cm⁻² resolution is reached, and thermal conductivity measurements on polymer films as thin as 25 μm, with a wide range of accessible values from ~0.1 to 0.8 W K⁻¹ m⁻¹.

INTRODUCTION

Printed electronics is widely regarded as the route toward cheap, large-area, flexible, and easily scalable electronics.¹ Carbon-based circuit boards including transistors, capacitors, and resistors can now be printed with competitive performance. However, the extra temperature sensitivity resulting from their organic nature has made thermal management in these components all the more critical. Temperature sensors, which are printable on flexible substrates, can help monitor the temperature and heat flux inside organic circuit-boards and identify the potentially overheating areas. In addition to thermal management, these printable sensors can be used in wearable electronics for healthcare, where the monitoring of human body temperature is especially important as minimal changes lead to drastic effects on the metabolism.² Printed temperature sensors can even be used to quantify the performance of newly developed cooling devices such as thermoelectric or electrocaloric coolers,³ where direct in situ measurements of heat flux are needed. Due to these growing interest, the number of publications on flexible sensors has increased 3-fold over the last ten years.^{2,4–7} These flexible temperature sensors are mostly resistance temperature sensors, which rely on the strong temperature dependence of their electrical resistance. Typical flexible resistance temperature sensors use expensive inorganic metals, such as platinum or

gold, either as coating produced by deposition, evaporation or sputtering,^{8–10} or through printable inks containing metallic micro- or nano-objects (particles, wires, ...).^{11,12} Also used are insulating or semiconducting polymer matrices, such as poly(3,4-ethylene dioxythiophene)-poly(styrenesulfonate), in which thermally conducting loads, such as carbon black or carbon nanotubes, have been added.^{2,13} Printing techniques including screen-printing^{14,15} or inkjet printing^{16–18} can then be used to print various materials on flexible substrates.

Here, we report a fully printed resistance temperature sensor using silver particles. Silver inks offer a cheap (<10 \$ g⁻¹) alternative to expensive commercial platinum sensors and are compatible with the easily scalable screen-printing technique. Printed silver-based sensors have been reported to display good performance, with sensitivity values close to that of platinum-based sensors (>0.2% °C),^{2,8,19} building on the sensitivity of bulk silver (0.38% °C).⁷ However, they are still

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unable to reach the state-of-the-art temperature resolutions ($<0.1\text{ }^{\circ}\text{C}$) recently reported in sensors with more complex architectures, such as designs using reduced graphene oxide.^{20,21} By combining a highly sensitive micropattern obtained using scalable screen-printing with an advanced signal treatment, we reach a $10\text{ m}^{\circ}\text{C}$ thermal resolution. As a result, the temperature sensors can be used for in situ heat flux measurements, with a 2 mW cm^{-2} resolution, and thermal conductivity measurements on polymer films as thin as $25\text{ }\mu\text{m}$.

RESULTS AND DISCUSSION

The temperature sensors consist of a conductive line of silver particles arranged to maximize the length of the line, and thus the resistance, in the sensing area by using a meandering pattern, as shown in Figure 1a. The temperature sensing mechanisms of the sensor relies on the increase of the electronic resistivity of a metallic conductor (silver here), with increasing temperature. This results from the increase of molecular vibrations, which hinder the movement of free electrons. The silver line width was set to $300\text{ }\mu\text{m}$, while the overall sensing area was 2.3 cm^{-2} . Wider silver pads were printed outside the sensing area for the electrical connection to the data acquisition system. A low-cost commercial silver ink was screen-printed in the desired pattern (printing protocol in Supporting Information). This design of temperature sensor was printed successfully on various flexible substrates, including poly(ethylene terephthalate) (PET) as shown in Figure 1b, as well as poly(ethylene naphthalate) (PEN) and polyimide (PI). A homogeneous silver particle conductive line with a width of $300\text{ }\mu\text{m}$ was obtained after only one run of screen-printing, as confirmed by the scanning electron microscope micrograph displayed in Figure 1c. In addition to its compatibility with roll-to-roll processes, and thus its scalability, screen-printing is a fast process, which allows the printing of the pattern in $\sim 1\text{ min}$ in a sheet-to-sheet process. With a large screen, several devices can be printed simultaneously without increasing the production duration, which is a great advantage compared to other technologies such as inkjet, which generally have a lower throughput.

The resistance of the temperature sensors (R_2) was characterized using a voltage divider represented schematically in the inset of Figure 2a. The temperature sensor is placed in series with a resistor of known resistance (R_1) and the output voltage U_2 is measured for a given input voltage U so that

$$R_2 = \frac{U_2 R_1}{U - U_2}$$

U_2 was recorded with a 16-bit analog-to-numeric converter. The evolution with temperature of the resistance of the temperature sensor is displayed in Figure 2a from 22 to $40\text{ }^{\circ}\text{C}$, which includes the temperature range of interest for body temperature monitoring ($36\text{--}40\text{ }^{\circ}\text{C}$).⁴ It shows that a $1\text{ }^{\circ}\text{C}$ change of temperature corresponds to a change of $287\text{ m}\Omega$ in resistance (dR/dT). This resolution is better than recently reported values for a similar system of $dR/dT = 109\text{ m}\Omega\text{ }^{\circ}\text{C}^{-1}$, due to our decreased silver line width of $300\text{ }\mu\text{m}$, compared to $500\text{ }\mu\text{m}$ used by Liew et al.¹⁸ This resolution could be further enhanced by increasing the overall surface area of the temperature sensors, which would increase the overall length of the silver resistance temperature sensor. However, we decided to use a small surface area (2.3 cm^{-2}) for a wider variety of possible applications. Further improvement could

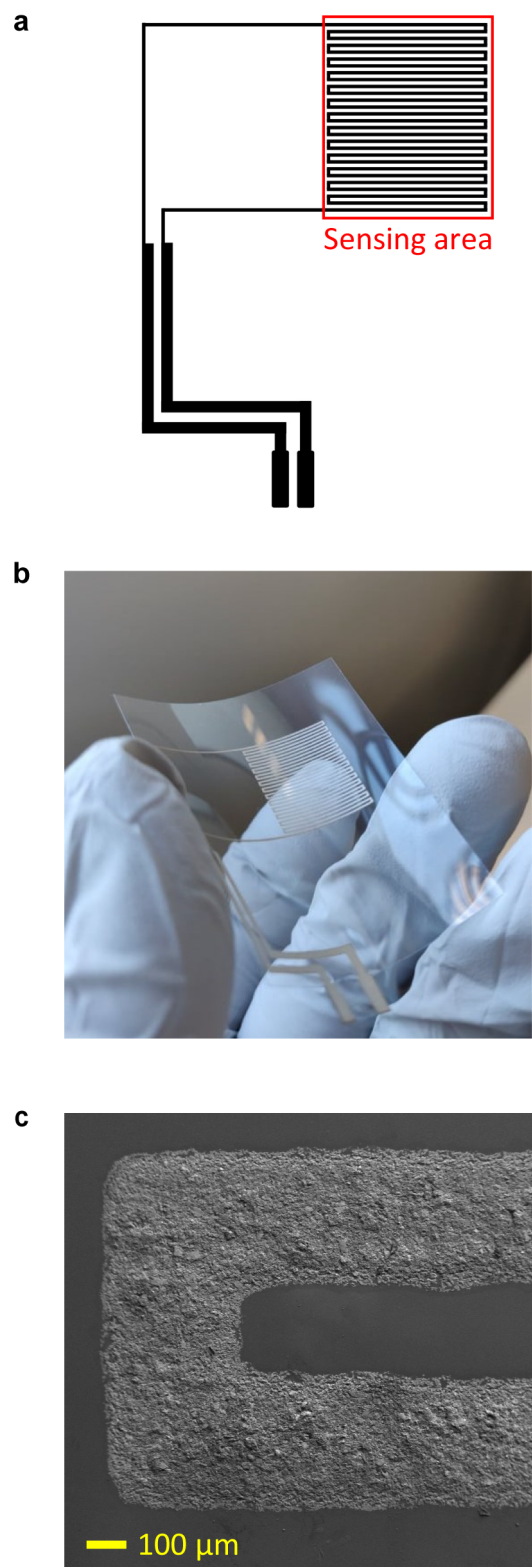


Figure 1. (a) Schematic representation of the temperature sensor. (b) Optical image of the temperature sensor screen-printed on a flexible PET substrate. (c) Scanning electron microscope micrograph of the silver particle conductive line of the printed temperature sensor (magnification $\times 75$).

also be obtained with thinner resistance temperature sensors, but decreasing the line width $<300\text{ }\mu\text{m}$ proved detrimental to

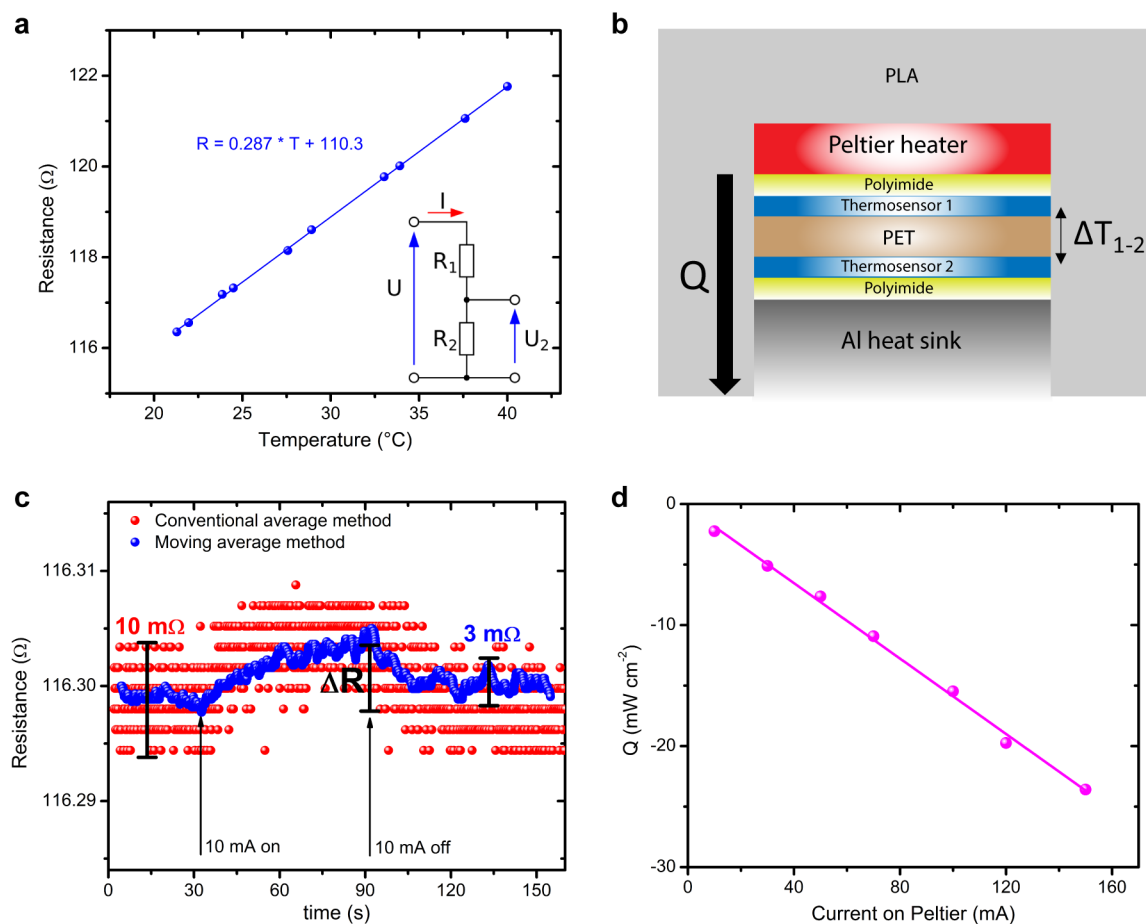


Figure 2. (a) Resistance versus temperature measured for the printed resistance temperature sensor. The inset shows the voltage divider used for the measurement of the resistance R_2 . (b) Schematic representation of the setup designed to study the influence of the temperature sensor's resistance resolution on its temperature and heat-flux measurement capabilities. (c) Comparison of the effect of the averaging method on the resolution of the resistance measurement. (d) Heat flux measured as a function of various current values applied to the Peltier element from 10 to 150 mA.

the quality of the patterns produced by screen-printing. Alternatively, inkjet printing could produce thinner lines and be used to print similar types of silver resistance temperature sensors.^{17,22,23} However, screen-printed was favored due to its aforementioned higher fabrication speed and scalability.

The most common tool to quantify the resolution of such sensors is the temperature coefficient of resistance (TCR), also sometimes referred to as temperature sensitivity (S), defined as

$$\text{TCR} = \frac{1}{R_0} \frac{R - R_0}{T - T_0}$$

$\text{TCR} = 0.25\% \text{ } ^\circ\text{C}^{-1}$ was measured for our printed temperature sensor with reference temperature $T_0 = 22 \text{ } ^\circ\text{C}$. This is comparable to the TCR of reference platinum temperature sensors ($\text{TCR} = 0.2\text{--}0.4\% \text{ } ^\circ\text{C}^{-1}$), while offering a much cheaper alternative.^{2,8} While the potential oxidation of silver could eventually result in poor long-term stability of the sensors; their use for in situ measurements, as proposed here, or healthcare applications, would require them to be buried inside a larger sealed structure or encapsulated, thus preventing their oxidation.

Building on this promising TCR, a high thermal resolution of $10 \text{ m}^\circ\text{C}$ was targeted for this printed temperature sensor. Instead of further increasing the TCR, this was achieved by reaching a resistance resolution of $3 \text{ m}\Omega$ (as $dR/dT \sim 0.3 \text{ }\Omega$

$^\circ\text{C}^{-1}$), through the optimization of U and R_1 for the advanced data acquisition of the 16 bit analog-to-numeric converter as well as the use of a moving average signal “treatment”.

A small setup was designed to study the influence of the temperature sensor's resistance resolution on its temperature and heat-flux measurement capabilities. Two temperature sensors were screen-printed on a $75\text{-}\mu\text{m}$ -thick PET substrate, with one on each side and overlapping sensing areas. They were then inserted between a Peltier heater and a thick (6 mm) aluminum heat sink, using $50\text{-}\mu\text{m}$ -thick polyimide tape for electrical insulation between each layer, as shown in Figure 2b. The whole stack was then thermally insulated using a poly(lactic acid) (PLA) box tailor-made by 3D-printing.

The resistance resolution was first decreased down to $10 \text{ m}\Omega$ by stabilizing U and optimizing R_1 . U was applied using a constant voltage source with a voltage stability down to a few microvolts in order to avoid additional parasitic noise from the voltage source during the resistance data acquisition. The reference resistance R_1 was then set to $1580 \text{ }\Omega$ in order to keep it high enough for the overall current flowing through the circuit to remain below 10 mA (highest current supported by the constant voltage source), but also as low as possible to reach the highest resolution available with our choice of analog-to-digital converter (see Supporting Information for more details, Figure S1).

The resistance resolution was then further decreased down to 3 m Ω by using a moving average method instead of a conventional average method. In both cases each data point corresponds to the average measurement over n points. However, in a conventional average method, the first data point is displayed after the n th measurement, as shown in in Figure S2a for $n = 6$, then the second data point is displayed after the $2n$ th measurement. This method significantly limits the measurement frequency, which is then divided by n . As a result, only 2 points are displayed after having measured 12 values of resistance in Figure S2a. When using a moving average method, the averaging is done over the n previously measured resistance values, instead of having to measure n new values before averaging. This allows to free the measurement frequency from n , which can then be set to a much higher value, even for high frequency. Figure S2b, shows that, for $n = 6$, 7 data points are displayed after having measured 12 values of resistance with a moving average method, compared to only two data points in the case of a conventional average method. The influence of the averaging method on the resistance resolution is shown in Figure 2c, where the time dependence of the resistance of the temperature sensor 2 is measured at 10 Hz with both methods. With the conventional averaging method, each data points corresponds to the average over 6 resistance values ($n = 6$), which result in the previously mentioned resistance resolution of 10 m Ω . However, using the moving average method, it was possible to increase 16-fold the number of resistance values for each data point ($n = 96$), while keeping the same measurement frequency. As a result, the resistance resolution was decreased from 10 to 3 m Ω . This resistance resolution corresponds to the targeted temperature resolution of 10 m $^{\circ}\text{C}$ with the printed temperature sensor. To illustrate the impact of this resolution enhancement, a current generating a small heat flux was applied to the Peltier heater at 30 s and then removed 60 s later. As the temperature sensor 2 is the furthest away from the Peltier element, it withstands the smallest temperature-induced change in resistance. Figure 2c shows that the resolution provided by the conventional averaging method does not allow for an accurate estimation of the temperature-induced resistance change, only a vague trend of increasing resistance can be observed. However, when using the moving average method, the significantly increased resistance resolution allows for an accurate measurement of the change in resistance, even for such a small current applied to the Peltier heater. While better TCR values have been achieved recently, especially in reduced graphene oxide (rGO) based sensors,^{20,21,24} a resolution of 10 m $^{\circ}\text{C}$ ranks among the best reported performance in flexible sensors, as shown in Table 1.

The scalability and the flexibility of these printed sensors, in which the performances were found to be maintained upon bending at various angles (Figure S3), provide them with a wide variety of potential applications. In addition to temperature data, using two temperature sensors with overlapping area and a substrate with known thermal conductivity gives access to the heat flux Q through the substrate as

$$Q = \frac{\Delta T_{1-2}}{R_{PET}} = \frac{AK_{PET}\Delta T_{1-2}}{d_{PET}}$$

where ΔT_{1-2} is the temperature difference between the temperature sensor 1 and the temperature sensor 2 and R_{PET} is the thermal resistance of the reference PET substrate, with d_{PET} the thickness and A the active surface area. The thermal

Table 1. Comparison of the Performance Indicators of our Printed Sensors with Reported Temperature Sensors Using Different Sensing Materials

sensing material	test range ($^{\circ}\text{C}$)	TCR (% $^{\circ}\text{C}^{-1}$)	resolution ($^{\circ}\text{C}$)	flexibility	ref
Ag	22–40	0.25	0.01	yes	this work
Ag	20–60	0.22		yes	17
Ag	30–100	0.10		yes	18
Pt	25–115	0.21		yes	8
Pt	20–120	0.32		yes	19
rGO	25–45	1.3	0.1	yes	20
rGO	30–80	1.34	0.2	yes	21
rGO	25–43	0.62		yes	24
graphene	20–50	1.34	5	yes	25
graphene	30–80	0.62	5	no	26

conductivity of the reference PET substrate was measured using the transient plane heat source (hot disc) method²⁷ (ISO 22007-2) and found to be $K_{PET} = 0.147 \text{ W K}^{-1} \text{ m}^{-1}$. Figure 2d displays the heat flux measured as a function of various current values applied to the Peltier element. It shows that the temperature resolution of the temperature sensors allowed the measurement of heat fluxes as small as 2 mW cm^{-2} . This range of accessible heat flux values opens the door to a wide variety of potential applications. For instance, Ma et al. recently reported a flexible electrocaloric cooler based on self-actuation,³ in which the smallest heat flux that they required to measure was $\sim 2 \text{ mW cm}^{-2}$. Although their design and its performance were promising, they added that the bulky heat flux sensor that they used for the measurement had a detrimental effect on the actuation. Printing our high-resolution temperature sensor directly on the flexible cooler would bypass this issue and provide the actual cooling performance of the device.

This technology could also be used to measure the thermal conductivity of polymer films with a steady-state method. It can be achieved by printing temperature sensors on the polymer sample of interest and mounting it in series with a material of known thermal conductivity, on which another set of temperature sensors has been printed, and then flowing heat through the multistacks, as shown in Figure 3a, where PET is used as the reference material. The temperature gradients for a given heat flux are then measured. This approach is thus similar to the commonly used comparative cut-bar technique.²⁸ Figure 3b shows the temperature gradients through a 75- μm -thick DuPont Kapton MT sample and the 75- μm -thick reference PET, ΔT_{3-4} and ΔT_{1-2} respectively, when a 37 mW cm^{-2} heat flux is applied with the Peltier. After 300 s the steady-state is established and the heat flux is constant so that

$$Q = Q_{1-2} = Q_{3-4},$$

the thermal conductivity of the sample can thus be extracted as

$$\frac{K_{PET}\Delta T_{1-2}}{d_{PET}} = \frac{K_{sample}\Delta T_{3-4}}{d_{sample}}$$

Under these conditions, the thermal conductivity of Kapton MT, K_{MT} , was found to be $0.46 \pm 0.03 \text{ W K}^{-1} \text{ m}^{-1}$, which is good agreement with the values reported by the manufacturer for this polymer of $0.46 \text{ W K}^{-1} \text{ m}^{-1}$. Several commercial polyimides with various thermal conductivities were characterized with this method and the results were compared with

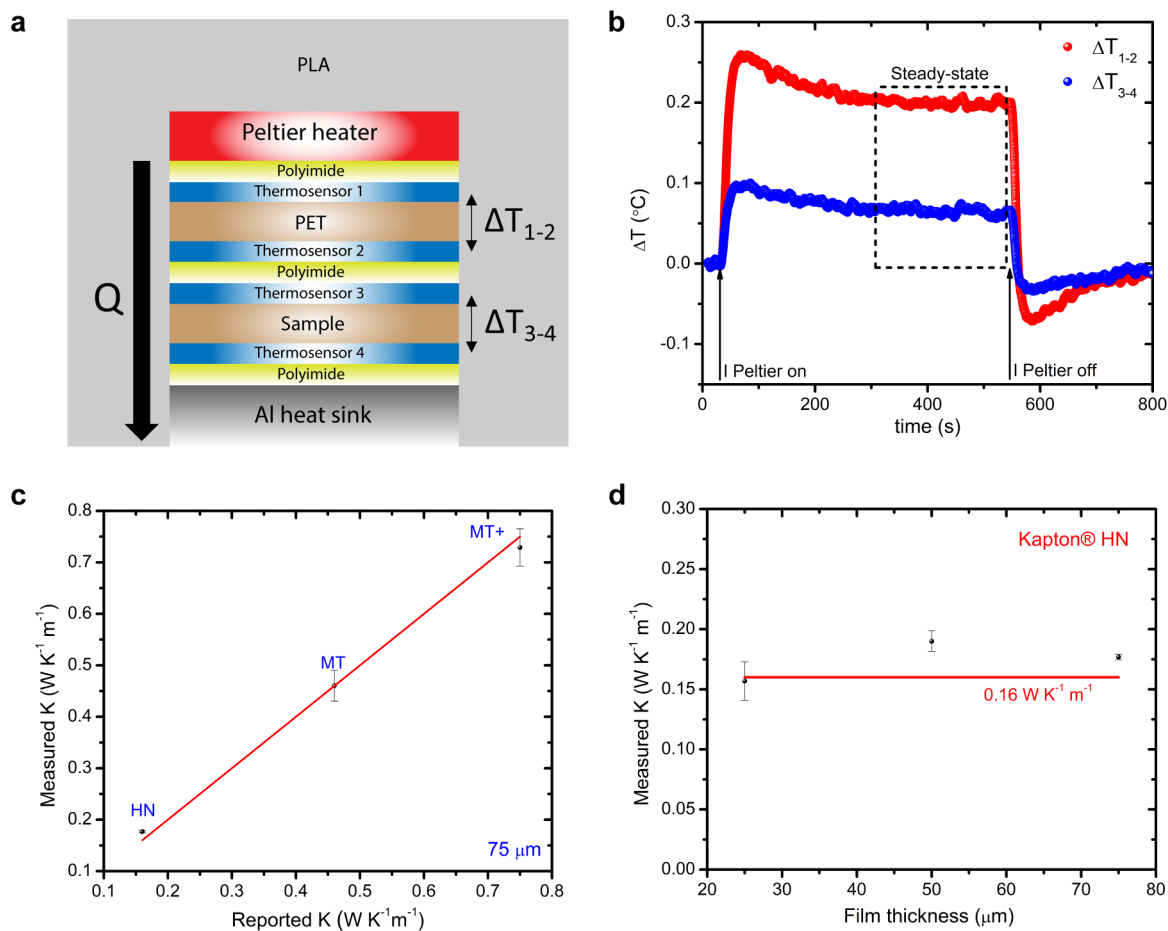


Figure 3. (a) Schematic representation of the setup designed to measure the thermal conductivity of a polymer film using four printed temperature sensors. (b) Temperature gradients measured through a 75- μm -thick Kapton MT sample and the 75- μm -thick reference PET, ΔT_{3-4} and ΔT_{1-2} respectively, when a 37 $mW cm^{-2}$ heat flux is applied with the Peltier heater at 22 °C. (c) Measured thermal conductivity versus reported values for various 75- μm -thick polyimide films: Kapton HN, Kapton MT, and Kapton MT+. The red line has a slope of 1. (d) Measured thermal conductivity versus film thickness for polyimide Kapton HN.

values provided by the manufacturers. 75- μm -thick films of DuPont Kapton HN, Kapton MT, and Kapton MT+ were selected, with reported thermal conductivities of 0.16, 0.46, and 0.75 $W K^{-1} m^{-1}$ respectively.²⁹ A good agreement was found for all these polymers, as shown in Figure 3c. As expected the error bar increased with the thermal conductivity of the sample, from $\pm 0.01 W K^{-1} m^{-1}$ for Kapton HN to $\pm 0.04 W K^{-1} m^{-1}$ for Kapton MT+. This is due to the lower temperature gradient induced through this more conductive materials, which was closer to the detection limit of the temperature sensors for Kapton MT+. However, the quality of the measurement was assured by adapting the value of the heat flux applied during the measurement, from 20 to 60 $mW cm^{-2}$ depending on the thermal resistance of the sample, in order to maintain $\Delta T_{3-4} > 0.06$ °C, which was thus six times higher than the thermal resolution of 0.01 °C.

This method was also used to measure the thermal conductivity of films with various thicknesses. Satisfactory results were obtained for films as thin as 25 μm with a measured value for Kapton HN of $0.16 \pm 0.02 W K^{-1} m^{-1}$ compared with an average reported value of 0.16 $W K^{-1} m^{-1}$, as shown in Figure 3d, where the comparison is given for three different thicknesses (25, 50, and 75 μm) of Kapton HN. These results further highlight the high quality and versatility

of the measurements performed with these printed temperature sensors.³⁰

CONCLUSIONS

In summary, we have designed and produced a temperature sensor, which can be printed on flexible substrates including PET, PEN, and PI. The choice of a silver particle ink offers an alternative to expensive commercial platinum sensors, with the added benefit of the use of easily scalable screen-printing. By combining a highly sensitive silver micropattern with an advanced signal treatment based on a moving average data acquisition, we were able to reach a 10 $m^\circ C$ thermal resolution. By designing an adapted testing system with 3D-printing, we have demonstrated the versatility of our temperature sensor and its potential for a wide variety of applications, including in situ heat flux measurements, where a 2 $mW cm^{-2}$ resolution was reached, and thermal conductivity measurements on polymer films as thin as 25 μm , with a wide range of accessible values from ~ 0.1 to 0.8 $W K^{-1} m^{-1}$.

ASSOCIATED CONTENT

Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsomega.2c07590>.

Experimental details, a detailed description of the electronic setup used to measure the resistance values of up to four printed resistance temperature sensors, a description of the averaging method used for the measurements, and measurements of the resistance of the flexible printed temperature sensors upon bending with different curvature angles (PDF)

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Notes

The authors declare no competing financial interest.

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