A Comparison of Relative Seebeck Coefficients for Screen Printed Flexible Thermocouples Using Commercially Available Conductive Inks

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***Abstract*— Thermocouples prepared on flexible substrates using additive manufacturing techniques can provide a path forward to strain independent temperature sensors for applications in a variety of wearable devices. An evaluation of 3 conductive ink pairings (C/Ag, Ag/Ni, C/Ni) fabricated using a screen-printed process is presented. The relative Seebeck coefficients of each ink pair was measured to be 9µV/°C +/- 2µV, 16 µV/°C+/-2µV and 27µV/°C +/-6µV respectively. Response time of the sensors were also evaluated to characterize the capability of real-time measurement with these devices.**

Keywords—flexible sensors; screen printing; flexible hybrid electronics; temperature sensor; Seebeck effect

I. Introduction

Rising global temperatures and increased frequency of heat wave events have given rise to higher risks of heat stress, particularly in vulnerable populations such as the elderly or outdoor workers[1]. Development of flexible sensing systems to monitor key physiological symptoms of heat stress has the potential to enable comfortable, continuous monitoring and provide early warning to reduce exertion or seek cooler environments.

Core temperature is a key factor in heat stress risk. Conventional methods of core temperature measurement, such as oral or rectal measurement, are invasive and unsuitable for daily use [2]. Distributed measurement of a body's surface temperature can be used to predict core temperature but require multiple temperature sensors placed at various points across the body [2]. Screen printed flexible sensors provide a low-cost, manufacturable solution. Resistive temperature devices are often used but suffer from strain dependance due to the flexible form factor. Thermocouples do not exhibit a strain response and therefore offer a desirable approach for flexible sensing. Printed thermocouples have been effectively demonstrated to provide strain insensitive measurements [3]. However, most of these approaches utilize laboratory developed ink and inkjet printers, or other fabrication techniques which are not preferable for manufacturability of flexible devices [3]–[5]. To successfully bridge the gap between laboratory development and widespread commercialization, identifying cost effective manufacturing solutions for such devices is necessary. To that end, we present a screen-printed fabrication process using commercially available inks which improves manufacturability, and can be integrated seamlessly into larger printed flexible electronics sensing systems.

Thermocouples utilize the Seebeck effect, a thermoelectric property by which a temperature difference across a conductor induces an electromotive force inside the conductor [6]. This force can be measured as a voltage differential across the

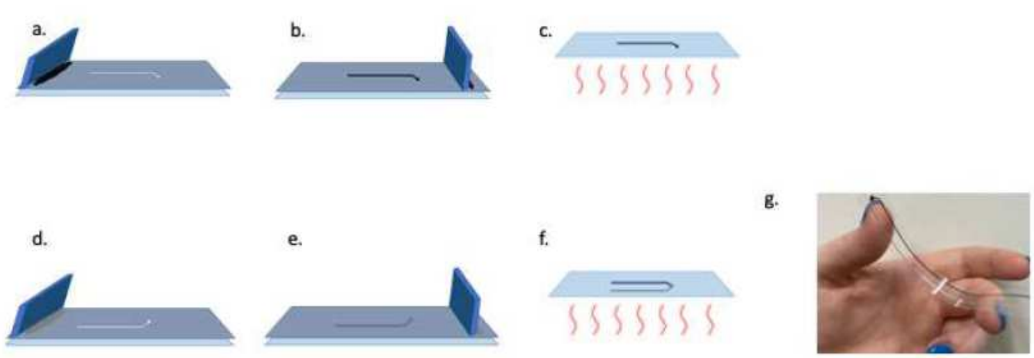

Fig. 1. Process flow of the screen printed thermocouple fabrication. **a.** Ink is applied to screen for first layer **b.** Squeegee presses ink through the screen pattern onto the substrate below **c.** First print layer is thermally cured to ink manufacturer's specifications. **d.-f.** Screen printing process is repeated for second ink layer. **g.** Image of printed thermocouple sample.

junction of two dissimilar materials exposed to the same temperature difference. In the working temperature range of a thermocouple, there is a linear relationship between the voltage differential across the junction and temperature, measured as

$$V_{AB} = \alpha_{AB} \cdot \delta T \quad (1)$$

where V_{AB} is the measured voltage potential, δT is the temperature gradient across the conductors, and α_{AB} is the relative Seebeck coefficient of the two conductors. The relative Seebeck coefficient is defined as

$$\alpha_{AB} = \alpha_A - \alpha_B \quad (2)$$

When the reference junction is held at a known steady temperature, typically 0°C, δT can be measured as the real temperature of the system[5]. The work presented here evaluates the relative Seebeck coefficient of three conductor pairs screen printed using commercially available inks, C/Ag, Ni/Ag, and C/Ni, across a temperature range relevant to human body temperature. The end result will be to look at the viability of these sensors for use in wearable devices.

II. METHODS

A. Fabrication

Polyethylene terephthalate (PET) substrates were prepared by cleaning the surface with isopropyl alcohol. A two layer design was fabricated by screen printing traces of commercially available conductive inks purchased from Nagase ChemTex: Silver ink CI-1036, Carbon ink CI-2051, and Nickel ink CI-5001. Fig. 1 illustrates the printing process of the samples. The first ink layer was printed and thermally cured according to the ink manufacturer's specifications, then the second ink layer was printed on top, forming the junction. The sample was then cured a second time.

Samples were prepared for testing using the ice point reference method by attaching two leads of the same material together to form a system with two conductor junctions. Wires of identical length were attached to the remaining sample leads for probe measurement. A layer of cellophane tape was applied to the surface of the sample to prevent oxidation due to air and moisture exposure.

B. Testing

The voltage response of each sample was measured using an ice bath setup to maintain a stable reference temperature of 0°C [6]. A Peltier heater was used as the heat source to allow dynamic control of the temperature at the measurement junction. A layer of thermal grease was applied to the surface of the heater, and the measurement junction was placed on top. The test setup was covered to minimize the effect of environmental air currents causing convective cooling of the sample. The voltage across the sample was measured using a digital multimeter (Agilent 34980A). The voltage applied to the heater was linearly increased to output temperatures in the range of interest, 24-40°C using a DC power supply (Keithley 2230-G). Reference measurements of both the ice bath and the heater were taken throughout testing using commercial type K thermocouples.

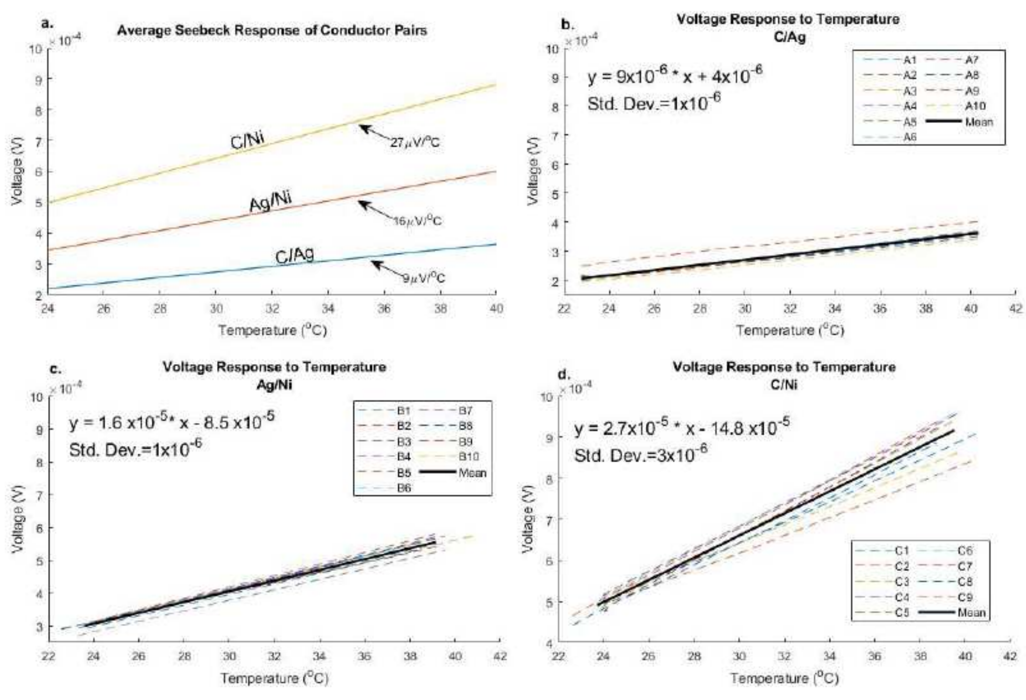

Fig. 2. **a**. Average relative Seebeck coefficient of C/Ag, Ag/Ni, and C/Ni samples. C/Ni exhibits the largest relative Seebeck coefficient at 27μV/°C. **b.** Voltage response of C/Ag samples. Samples show a mean relative Seebeck coefficient of 9 μV/°C with some variability in y-intercept. **c.** Voltage response of Ag/Ni samples. Samples show a mean relative Seebeck coefficient of 16 μV/°C. **d.** Voltage response of C/Ni. Samples show a mean relative Seebeck coefficient of 27 μV/°C, with a higher variability than Ni/Ag or C/Ag pairs.

III. Results and Discussion

Ten samples of each thermocouple were fabricated and tested to determine the average relative Seebeck coefficient of the conductor pair. Samples were placed in the test setup and measured at 15 points between 22 and 40°C. A linear fit of each sample was obtained to interpolate the behavior of the sample between measurement points, and then averaged to find the generalized behavior of the conductor pair[7]. Fig. 2 presents the measurement data and mean relative Seebeck coefficient for each conductor pair.

Each conductor pair demonstrates a measurably different relative Seebeck coefficient, with C/Ni presenting the highest coefficient. Table 1 summarizes the results of this work compared with other printed thermocouples demonstrated in literature. Of the reported results, the C/Ni thermocouple presented in this work is among the highest relative Seebeck coefficient reported using commercially available inks. For thermocouples fabricated with similar materials, the presented devices demonstrate comparable results[3], [4]. The inks chosen in this study have been sourced from different companies than previous literature, each with their own proprietary formulation. Because the printed inks are not bulk materials, some variability can be expected due to differences in ink formulation and particle size between different ink manufacturers [8]. Additionally, particle dispersion and the addition of binders within the ink layers may vary between samples, which could result in the observed variation of relative Seebeck coefficient. Optimization of relative Seebeck coefficients may be possible by further investigating the specific formulation of commercial inks to understand its effect on relative Seebeck response.

TABLE I. Relative Seebeck Coefficients of Printed Flexible Thermocouples

Source	Materials Used	Commercially Available?	Relative Seebeck Coefficient (μV/°C)
This work	C/Ag Ag/Ni C/Ni	Yes Yes Yes	9 +/- 2 16 +/- 2 27 +/-6
[3]	SWCNT/MWCNT SWCNT/AgNW MWCNT/AgNW	No No No	24.36 37.63 9.93
[9]	C/Ag	Yes	Calculated Seebeck coefficient not disclosed.
[4]	Ag/Ni	Yes (Ni ink still under development)	20
[5]	Carbon black (Multiple Inks)	Yes	4.29 5.24 1.13

The response time of the samples was tested by submerging each sample in a water bath held at 40°C. Voltage output of the sample was measured for the duration of testing. Fig. 3 demonstrates the response time of an C/Ag sample.

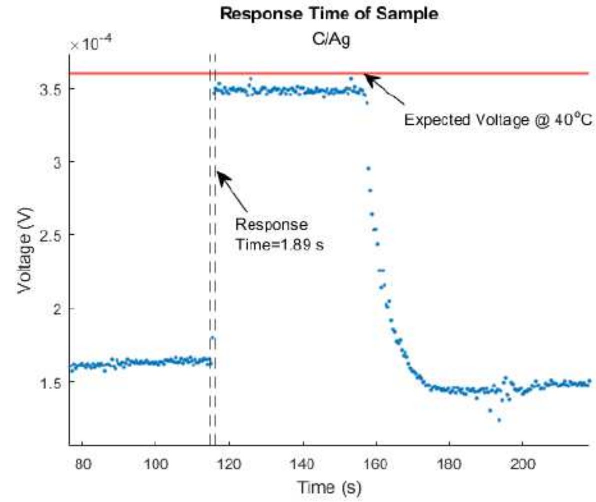

Fig. 3. Response time of C/Ag thermocouple sample when submerged in 40°C water bath. Voltage response reaches stable output at 1.89 s.

Response times of the 3 conductor pairs ranged from 1.89 – 2.83s, indicating good compatibility with real time measurement of body surface temperature for prolonged durations. Response time of thermocouples has been shown to be dependent on the thermal mass of the junction point and is therefore affected by the thickness of the printed ink layers[3]. The inks used in this study have an expected print thickness ranging between 7-17μm, however optimization of printing parameters is necessary to achieve minimum print thickness consistently.

IV. Conclusion

Flexible thermocouples were screen printed using three commercially available inks and their respective relative Seebeck coefficients were evaluated. Each ink pair demonstrated a unique relative Seebeck coefficient with minimal variation between samples. The C/Ni sample exhibited a relative Seebeck coefficient among the highest reported for screen printed thermocouples using commercially available inks. Response times of the sensors were evaluated and found to be less than 3 seconds for each ink pair. These sensors demonstrate strong potential for use in highly manufacturable, low cost wearable sensing systems. Further work will focus on optimization of Seebeck response by investigating the effect of ink formulation on relative Seebeck coefficients of devices, and identifying optimal printing parameters for ensuring low variability during high volume manufacturing. Additionally, the effect of strain on Seebeck response will be investigated to further confirm the sensors compatibility with wearable devices.

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