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Printed thick-film thermocouple sensors

S. Duby, B.J. Ramsey and D.J. Harrison

Thermocouples have been printed using thick-film processes, which demonstrate a repeatable and stable reaction to temperature and a potential difference per degree temperature change comparable to conventional commercial couples. The potential for this novel approach is to offer a rapid, low-cost, low-temperature manufacturing process for electrical temperature monitoring.

Introduction: The work reported in this Letter builds on previous work done at Brunel University on the printing of passive electronic components [1, 2], from simple capacitors and resistors, to more complex integrated filter structures, microwave circuit components and humidity sensors [3]. Printed thermocouples have been manufactured using a variety of specifically formulated thermoelectrically active inks (see Fig. 1). The printed *p*- and *n*-type structures have been thermoelectrically characterised, and their combination into a thermocouple configuration (see Fig. 2) has resulted in generated electromotive force (EMF) per degree temperature differences comparable to standard thermocouple configurations. Results also compare favourably with the thermoelectric performance of other work on novel thermocouple manufacture methods, including that reported by Wen and Chung [4], and Nogi *et al.* [5].

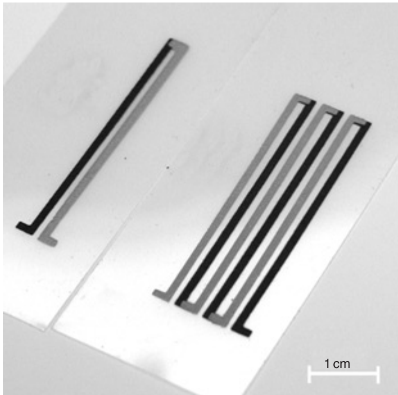

Fig. 1 *Printed thermocouples*

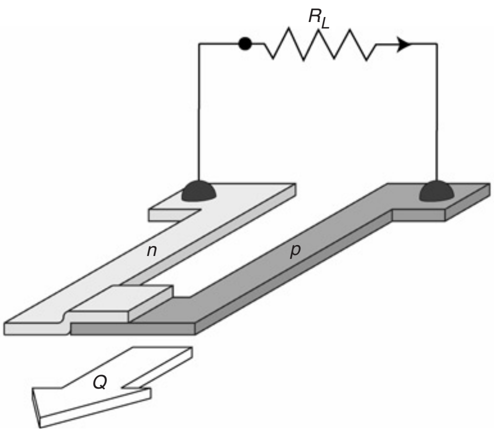

Fig. 2 *Principle of thermocouple manufacture and operation*

The inks developed for this process consist of metallic particulates suspended in a variety of polyester, cellulose and conductive polyanaline (Panipol M) resins. The principal functions of the resin vehicle are to provide suitable flow characteristics for the ink, to ensure that the final printed track has structural integrity and to ensure that the active particulate is adequately adhered to the polyester substrate. As great a particulate loading as is viable is desirable.

Method: Inks were all manufactured in-house in our ink labs using commercially sourced materials. The inks were formulated to conform to the required rheological criteria, screen printed and electrically assessed for thermoelectric suitability. The Seebeck coefficients of the samples were obtained using test apparatus that held the ends of the thermoelement or thermocouple at two stable and different temperatures (maintaining a ∂T of approximately 150°C) while measuring the generated EMF. The technique was in accordance with prior methods as described by Kayadanov and Ohno [6] and Yang *et al.* [7]. Results were obtained for printed samples of single legs containing various material combinations and solid samples of standard thermoelectric materials. Results were also obtained for the reaction of printed thermocouple configurations and standard thermocouples.

The apparatus was designed such that one end of the thermocouple was held between two air-cooled Peltier modules capable of maintaining a closely controllable temperature of down to −20°C, while the other was held between two flat-plate resistance heaters powered by a Zenith 240 V AC variable transformer capable of maintaining a stable temperature of between ambient and 170°C. The sample was only otherwise in contact with the air, ensuring that heat flow was predominately through the printed track with a negligible amount through the polyester substrate. Three mineral-insulated *k*-type thermocouples probes continually monitored the ambient, cold-junction and hot-junction temperatures through a Pico TC-08 eight-channel thermocouple data logger connected to a PC. The TC-08 data logger was capable of measuring absolute potential difference with a resolution of unitary microvolts and was also used to monitor the generated EMF of the sample. Results from the testing of standard thermocouple legs using this setup correlate with theoretical data (within 5% variance) [8], see Table 1.

Table 1: Correlation between experimental and theoretical Seebeck coefficients

Material	Theoretical (mV/°C)	Experimental (mV/°C)
NiCr wire	2.5×10^{-2}	2.4×10^{-2}
NiCr/CuNi (E-type)	6.8×10^{-2}	6.2×10^{-2}

Various additives have been included in the formulation that have provided significant improvements to the electrical properties. Wetting agents have been employed to separate the individual particles from one another and aid dispersion in the ink, as well as ensuring that a minimum amount of binder is needed to coat each individual particle completely. Other materials have been added to penetrate and destabilise passive oxide layers on the metallic particles.

Results: The thermoelectrically generated potential differences of the printed couples are comparable to those of conventional couples (see Table 2). The sum of the experimental results for Seebeck voltages of the separate thermoelements of a standard couple are in close accordance with the theoretical data for the complete couple.

Table 2: Published bulk material Seebeck coefficients compared to those of printed samples

Material	Bulk material theory (mV/°C)	Experimental (mV/°C) printed
NiCr	2.5×10^{-2}	9.37×10^{-3}
Fe	1.9×10^{-2}	4.96×10^{-3}
Ag	6.5×10^{-3}	2.87×10^{-3}
Ni	-1.5×10^{-2}	-1.03×10^{-2}
Ni/Panipol	N/A	-1.25×10^{-2}
NiCr/Ag	N/A	4.14×10^{-3}

Discussion: Results indicate that generated Seebeck EMF was more dependent on the type and quantity of the principal thermoelectric material used than on the intrinsic electrical resistivity of the material. The amount of thermoelectrically active material in the formulation, which in this instance is analogous to the particulate loading of the ink, was defined by the required physical characteristics of the ink.

Particulate loadings of 96% by volume in the dried deposited sample have been achieved. These provide a greater per degree potential difference, but the constraints of adequate adhesion mean that lower particulate loadings are more suitable. The inks were deposited onto a flexible polyester substrate so the binder system had to be compatible with this type of material.

Discrepancies between the maximum Seebeck voltage of printed samples and those of standard couples can be partially explained by the fact that the active material in a printed sample is in the form of particulate held in a non-conductive, non-thermoelectrically active matrix. This means that electrical conduction is a function of the efficiency of the tunnelling of electrons from one metallic particle to the next. Consequently, the formulation of the ink is of critical importance, as this inter-particulate conduction is dependent on a variety of factors including particulate leafing and the absence or presence of non-conductive oxide layers on the individual particles.

The reaction of the printed devices to temperature over the range 0–150°C appears typical for that expected from a standard temperature sensing thermocouple; stable and predictable. This is shown in Fig. 3, where printed couple ***a*** and printed couple ***b*** are, respectively, a *p*-type Ni/Panipol-based ink and *n*-type silver-based ink combination, and a *p*-type Ni/Panipol-based ink and nichrome-based ink combination. A preliminary assessment of the variation to be expected from manufacturing variables has been undertaken. The thermoelectric response from devices printed on different occasions and from different batches of ink has proved consistent with a standard deviation of <0.01.

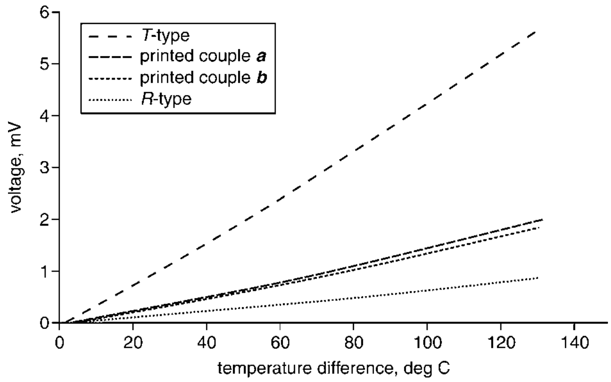

Fig. 3 *Generated Seebeck voltages of two printed and two standard thermocouples*

Conclusions: Thick-film manufacture processes have been employed to fabricate temperature sensors based on the principles of thermoelectricity. Separately developed *p*- and *n*-type materials have been used in conjunction in a thermocouple arrangement. Table 2 demonstrates the thermoelectric reaction of single legs. It can be seen that a combination of the two highest performing materials (*p*-type Ni/Panipol and *n*-type NiCr) will give a reaction of 22 μV/°C ($1.25 \times 10^2 + 9.37 \times 10^3 = 2.18 \times 10^2$), which is comparable to the reaction of conventional thermocouples. The experimental results indicate a potential for this approach to offer a rapid, low-cost, low-temperature manufacturing process for effective electrical temperature monitoring devices, overcoming the need to place and solder components and the requirement to 'fire' and laser trim ceramic systems.

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