Sensors and Actuators A 279 (2018) 84–89

Contents lists available at ScienceDirect

Sensors and Actuators A: Physical

journal homepage: www.elsevier.com/locate/sna

A fully spray processed embedded composite thermocouple for the use at high temperatures and harsh environments

Marcel Knoll*, Christina Offenzeller, Bernhard Jakoby, Wolfgang Hilber

Institute for Microelectronics and Microsensors, Johannes Kepler University, Altenbergerstrasse 69, 4040, Linz, Austria

ARTICLE INFO

Article history:
Received 9 January 2018
Received in revised form 27 March 2018
Accepted 5 June 2018
Available online 6 June 2018

Keywords:
Metal/organic thermocouple
Embedded transducer
Polymer based, organic coatings
Steel substrates

ABSTRACT

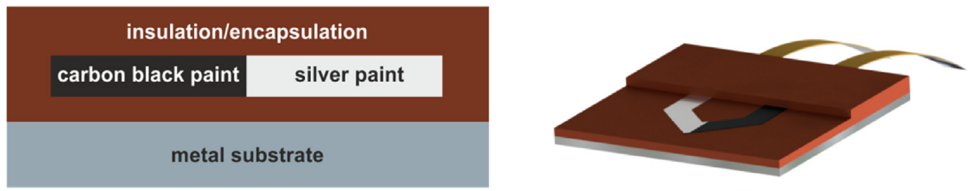
We present a fully spray processed polymer based temperature sensor which can be embedded in the organic coating of metallic machine components. The main aim of the sensor is to measure the temperature in such coatings. This could be used, e.g., in friction bearings for condition monitoring. For the present demonstrator, the sensor element is embedded on a steel substrate. Here, the sensing element is a thermocouple, which is made out of two conductive paints with carbon black and silver as organic or metallic filler particles, respectively. The used carbon black paint is custom-made and uses polyamide-imide as polymer binder which serves also as the polymer backbone of the insulation and the encapsulation layer. The commercially available silver paint is based on polyimide which yields the desired bond strength with respect to the insulation and encapsulation layer. The investigated thermocouples are characterized on a temperature test rig with and without top coating up to a junction temperature of 200 °C. Subsequently, the influence of prolonged heat treatment (in total 16.5 days at 200 °C) on the thermocouple sensitivity is investigated. Finally, the thermocouple cross-sensitivity study of the pressure influence on the temperature sensitivity is performed. First a test was made with pure pressure load of 40 MPa without a temperature gradient between junction and terminal. The final cross-sensitivity measurement was performed in a climatic chamber including a pressure test rig which is able to apply a load of 200 MPa at a maximum temperature of 141.5 °C.

1. Introduction

Thermocouples are very precise temperature sensors with a wide measuring span used in different application fields and realized in various embodiments [1–4]. This work represents an approach for the realization of an embedded, spray processed thermocouple. Thermocouples consist of two different conductors which are in contact at one end, which is referred to as junction. If a temperature gradient between junction and the open end (terminal) of the conductors is present, an output voltage is measureable between the open ends [5]. Thin film thermocouples are of interest for the realization of embedded temperature sensors, e.g., in an existing structure of metal that is coated with an organic paint. Embedding the sensor enables improvements in mechanical and chemical resistance but also may, e.g., facilitate a temperature measurement underneath a functional layer. Additional advantages of embedding the sensor are the faster response time and higher accuracy, as there is no air gap or adhesive layer between the sensor and the object to be characterized. Compared to other temperature transducing mechanisms like thermistors (which are resistors and therefore also act as strain gauges) and pyroelectrics, thermocouples typically show a pressure dependence only at very high pressures of several MPa [6]. This property makes thermocouples a promising temperature sensor for the use in harsh environments where mechanical strain or stress are present. Conventional methods for fabricating thin film thermocouples are vapor deposition [7] or sputtering [8,9]. In contrast to these expensive and time-consuming technologies, the spray process offers a low cost alternative, which allows a placement of the sensor on virtually any geometry of interest. The spray coating technology is a frequently used technique, e.g., for the fabrication of micro strain gauges [10], ion selective electrodes [11], or thin film gas sensors [12]. To our knowledge, polymer based thermocouples have not been reported so far. The investigated devices particularly feature polymeric electrodes and are implemented on a conducting metallic substrate. This design is demonstrated to be high temperature compatible (up to 200 °C) and moreover endures high pressures (up to 200 MPa). By virtue of this design, they are embeddable into existing coating systems of metallic machine parts. Thus these devices are feasible, e.g. for the implementation in friction bearings, where condition moni-

* Corresponding author.
E-mail address: marcel.knoll@jku.at (M. Knoll).

https://doi.org/10.1016/j.sna.2018.06.013



(a) (b)

Fig. 1. (a) Schematic geometry of the embedded thermocouple. The metal substrate is coated with a polyamide-imide insulation layer on which the thermocouple (consisting of the carbon black paint and the silver paint conducting tracks) is processed. The top layer is made out of the same material as the insulation (b) Representation of the embedded thermocouple. The polyimide connections (depicted in orange) are coated with the carbon black or the silver paint.

toring is an interesting feature which helps to prevent overstraining and destruction of the machine. Preliminary results associated with this work have been presented at the Eurosensors 2017 in Paris [13].

2. Materials and methods

2.1. Sensor architecture

The first layer to be processed on the metallic substrate is the insulation layer. Since the insulation as well as the encapsulation should withstand high temperatures and be mechanically as well as chemically stable, the polymer Rhodeftal 210 ES was chosen. Rhodeftal is a polyamide-imide mixture and suitable for temperatures up to 250 °C, where most polymers have already passed their melting point [14]. After the curing process Rhodeftal shows excellent mechanical and chemical stability. To achieve a good bond of the conducting layers on the insulation layer, paints with a similar polymer matrix are used. The first conductive paint is commercially available where the conductive filler particles are made of silver and the polymer binder is polyimide. The second conductive paint used is a custom-made carbon black paint with polyamide-imide as binder.

A schematic of the thermocouple layer architecture is depicted in Fig. 1(a). The substrate is spray coated with Rhodeftal. On top of the insulation layer the carbon black and the silver conductive paints are deposited using the airbrush. Finally, the conductive paints are top coated with Rhodeftal. Fig. 1(b) is a schematic representation of the whole thermocouple sensor. The orange strips in the sketch are polyimide strips (cut out of thin polyimide foil) coated with the according conductive paint. These strips are used later as electrical connections for the measurement.

2.2. Paint preparation and processing

For processing the insulation and the encapsulation layer, 100 g Rhodeftal is mixed with 120 g N-Methyl-2-pyrrolidone (NMP), 80 g p-Xylene and 0,3 g BYK 310 obtained from BYK Additives and Instruments. NMP and p-Xylene are solvents used for adapting the viscosity of the Rhodeftal for the spray coating process and BYK 310 is a surface additive used to reduce the surface tension of the paint.

The conductive silver paint KA 801 has been obtained from Dupont. The paint is diluted with Ethylacetate to obtain a spray processable solution. Thus the silver paint is mixed with Ethylacetate at a weight ratio of 1:1 using a magnetic stirrer.

For the preparation of the carbon black paint, 3 g of sieved carbon black are dispersed in a solvent mixture of 66 g p-Xylene, 57 g NMP, 0.08 g of BYK 310 and 0.132 g of the dispersing agent Nuosperse 196 obtained from Elementis Specialities. The mixture is stirred with an agitator for half an hour and afterwards placed in an ultrasonic bath for 8 h. The last step is the addition of the polymer binder, where 23.7 g of Rhodeftal 210 are dispersed in the solvent/carbon black mixture using a magnetic stirrer. After the mixing process the paint layers are applied in a spray process using an airbrush.

Prior to the insulation coating, the substrate is heated to 150 °C for 15–20 s. In total three coating steps are performed in order to achieve the desired thickness and uniformity. After the last coating step the paint has to be cured for 30 min at 250 °C. For the coating step of the carbon black and the silver paint, a structured adhesive mask is used. The insulated metal substrate is preheated for 20 s at 250 °C and coated with the carbon black paint afterwards. Subsequently, the paint is cured for 30 min at 250 °C. For the air brushing of the silver paint the substrate is placed on a hotplate at 200 °C for 20 s. After the coating step the sample is placed on a hotplate for 30 min at 200 °C. Thereafter polyimide strips, coated with the according conductive paint (where the coating process is the same as for the sensor layers), are connected to the silver or carbon black layer, respectively. The connection is done with a droplet of the according paint. Finally, the sample is top coated with Rhodeftal and cured for 30 min at 250 °C (first curing version) or with 200 °C (final curing version due to the silver paint stability). Fig. 2(a) shows the spray processed thermocouple without top coat.

3. Measurement and results

3.1. Influence of the top coating on the thermocouple sensitivity

The thermocouple was characterized in a custom made test rig which is depicted in Fig. 2(b). The thermocouple is mounted on an aluminum block, which includes a high power resistor as heating element. A PT-1000 thermistor is placed on top of the thermocouple junction to measure the junction temperature. The polyimide connections are fitted into a cooling box which contains a water tempered aluminum block, used for keeping the temperature of the electric terminals constant. The temperature of the aluminum block inside the box is measured with an additional temperature sensor.

During the characterization program the thermocouple junction is heated to 250 °C (if the first curing version was used) or 200 °C (if the final curing version of the top coating was used), respectively. Afterwards the cooling process, which takes 2 h and 20 min, begins. During the cooling process the output voltage and the temperature difference between the junction and the terminals are measured every second. Fig. 3 shows the recorded thermocouple voltage [mV] versus the temperature difference ΔT [°C] between the junction and the terminal for eight different measurements. The first four recorded curves show the temperature dependent voltage for a thermocouple without top coating (abbreviated in the legend with MEAS 1–4), and the next four curves depict the voltage-temperature dependency for the same thermocouple with top coating (abbreviated in the legend with MEAS 1–4 enc).

For each measurement the voltage-temperature response was found to be slightly nonlinear with increasing sensitivity for higher temperatures. The maximum output voltage of 4.32 mV is reached at a temperature difference of 225 °C.