proceedings

Eurosensors PARIS 2017

MDPI

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A Spray Processed Polymer-Based High Temperature Organic/Metal Thermocouple for Embedding in Organic Coatings of Steel Substrates †

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† Presented at the Eurosensors 2017 Conference, Paris, France, 3–6 September 2017.

Published: 28 August 2017

Abstract: In this work we present the realization of a fully spray processed embedded temperature sensor. The sensing element is a thermocouple made of conductive materials which are high temperature stable polymer based paints with organic or metallic filler particles. The thermocouple is embedded in an organic paint layer which shows, besides good high temperature properties up to 250 °C, excellent mechanical and chemical stability. The manufactured thermocouples are tested up to a junction temperature of 250 °C, first without and afterwards with an encapsulation layer. The measured output voltage for a terminal temperature of 25 °C and a junction temperature of 250 °C is in the region of 4.23 mV before and after top coating. Finally, a layer analysis of the encapsulated device is made using a cross-section polish.

Keywords: spray coated; thermocouple; embedded transducer; high temperature stable; polymer based; metal/organic; polymer composite

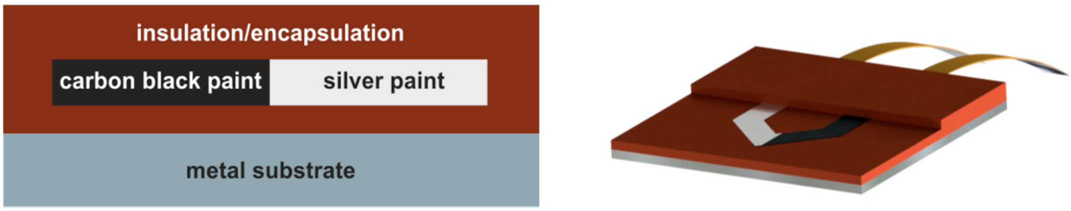
1. Introduction

Thermocouples are very precise temperature sensors with a wide measuring span used in different application fields and realized in various embodiments. This work is an approach for the realization of an embedded, spray processed thermocouple. Thermocouples consist of two different conductors which are in contact at one end, this end is called junction. If there is a temperature difference between the junction and the open end of the conductors, an output voltage between the open ends (terminal) is measurable. Thin film thermocouples are of interest for the realization of embedded temperature sensors, e.g., in an existing structure of metal coated with an organic paint to enable a better mechanical and chemical resistance of the sensor. An additional advantage of embedding the sensor arises from a faster response time and higher accuracy as there is no air gap or adhesive layer between the sensor and the object to be characterized. Conventional methods for producing thin film thermocouples are vapor deposition [1] or sputtering [2,3]. In contrast to these expensive and time-consuming technologies, the spray process offers a low cost alternative which makes it possible to place the sensor structure on almost every geometry of interest. The spray coating technology is a frequently used technique, which is, e.g., used for the production of micro strain gauges [4], ion selective electrodes [5], or thin film gas sensors [6].

2. Materials and Methods

2.1. Sensor Architecture

The first layer, which is processed on the metallic substrate, is the insulation layer. As the insulation as well as the encapsulation should withstand high temperatures and be mechanically and chemically stable, the chosen polymer is Rhodeftal 210 ES. Rhodeftal is a polyamide-imide mixture and suitable for high temperature applications up to 250 °C, where most polymers have already reached their melting point [7]. After the curing process Rhodeftal shows an excellent mechanical and chemical stability. To achieve a good bond of the conducting on the insulation layer, paints with a similar polymer matrix are used. The first conductive paint is a commercially available silver paint with a polyimide polymer binder, and the second conductive paint is a custom-made carbon black paint with a polyamide-imide polymer binder. A schematic of the thermocouple layer architecture is depicted in Figure 1a. The substrate is spray coated with Rhodeftal. On top of the insulation layer the carbon black and the silver conductive paints are processed using an airbrush. Finally, the conductive paints are top coated with Rhodeftal. Figure 1b is a schematic representation of the whole thermocouple sensor. The orange strips in the picture are poly imide strips coated with the according conductive paint. These strips are used later as electrical connections for the measurement.

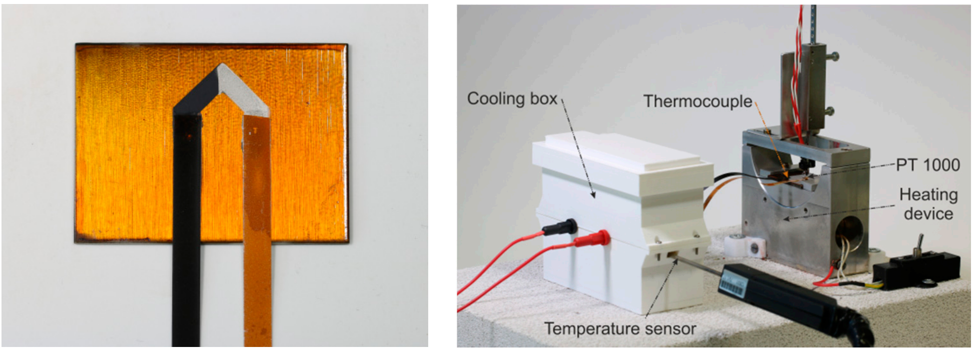


(**a**) (**b**)

Figure 1. (**a**) Schematic architecture 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) is processed. The top layer is made out of the same material as the insulation (**b**) schematic representation of the embedded thermocouple. The polyimide connections (depicted in orange) are coated with the carbon black or the silver paint.

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 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 for lowering 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 from Elementis Specialities. The mixture is stirred with an agitator for half an hour and afterwards is 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. The first production step is insulating the metal substrate with Rhodeftal. Prior to the coating step, the substrate is preheated to 150 °C for 15–20 s. In total three coating steps are performed for the insulation layer in order to achieve the desired thickness and homogeneity. 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. Finally, the sample is top coated with Rhodeftal and cured for 30 min at 250 °C. Figure 2a shows the spray processed thermocouple without top coat.



(**a**) (**b**)

Figure 2. (**a**) spray processed thermocouple on metal substrate with polyimide connections before top coating; (**b**) Thermocouple test rig consisting of the heating device, a PT 1000 (for measuring the thermocouple junction temperature), a cooling box with aluminum blocks (for keeping the connection temperature constant) and a temperature sensor to measure the connection temperature.

3. Measurement and Results

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

First the thermocouple junction is heated to 250 °C, afterwards the cooling process to room temperature starts. This process takes 2 h and 20 min. During the cooling process the output voltage and the temperature difference between the junction and the connection are measured every second. Figure 3 shows the recorded thermocouple voltage (mV) over the temperature difference ΔT (°C) between the junction and the connections 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 the voltage-temperature dependency for the same thermocouple with top coating (abbreviated in the legend with MEAS 1–4 enc). In each measurement the voltage-temperature response is nonlinear with increasing sensitivity for higher temperatures. The maximum output voltage of 4.32 mV is reached at 225 °C. The layer height of the conductive layers is determined using a cross-section polish. The resulting height is 3 μm for the silver, and 4 μm for the carbon black layer.

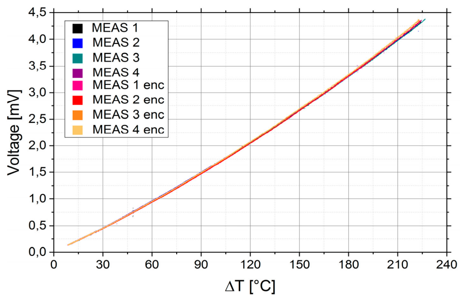


Figure 3. Graph of the recorded voltage over the temperature difference between junction and the connection for the uncoated (abbreviated with MEAS 1–4) and top coated (abbreviated with MEAS 1–4 enc) thermocouple.

4. Discussion and Conclusions

A spray processed metal/organic thermocouple was successfully fabricated. The determined maximum output voltage of 4.32 mV at a temperature difference of 225 °C is larger than that of other junction material combinations (0.39–10.45 mV, ΔT 200 °C) [8]. Furthermore, the thermocouple showed the same temperature-voltage behavior with and without encapsulation; therefore the top coating process does not affect the thermocouple's performance. Next steps are the investigations of the thermocouple pressure-temperature cross-sensitivity.

Acknowledgments: This work has been supported by the Linz Center of Mechatronics (LCM) in the framework of the Austrian COMET-K2 program.

Conflicts of Interest: The authors declare no conflict of interest.

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