Screen-printing Resistive Temperature Sensor development with sustainable carbon pastes

Leire Sanchez-Duenas, Cristina Monteserin, Estibaliz Gómez, Miren Blanco, Mikel Larrañaga, Estibaliz Aranzabe
Fundación Tekniker, Eibar, Spain.

Miryam Criado-Gonzalez, Rajat Rai
Polymat University of the Basque Country UPV/EHU, Donostia-San Sebastian, Spain

José Luis Vilas-Vilela
University of the Basque Country UPV/EHU, Leioa, Spain

Abstract—**During the last decades, increment of the demand of electronics and the programmed obsolescence have entailed a growth of the electronic waste. On this context, there is a need of developing new sustainable and/or recyclable electronic devices. Sustainability of new devices could be ensured by using biobased materials and avoiding those which are harmful for the environment. These new materials could be used to develop specific devices, as resistive temperature sensors deposited on flexible materials, for wearable devices. Resistive temperature sensors use their electrical resistivity variation with temperature to measure the environment temperature. The variation on the resistance of the sensor depends on the material they are composed of. On this work, resistive temperature sensors are printed by screen printing, with four different pastes: a silver based commercial paste and three carbon-based pastes, a commercial one and two sustainable developed pastes. The sensors were printed onto glass fiber substrate to ensure flexibility and infused with a resin to get a composite.**

Keywords—screen printing pastes, sustainability, resistive temperature sensor (RTD), silver paste, carbon paste, temperature coefficient resistance (TCR), PEDOT:Biopolymer, Multiwall carbon nanotubes (MWCNT), Porous Carbon Nanopowder, vanillin-derived Schiff base (VSB), epoxy resin.

I. INTRODUCTION

New electronic developments, specifically those developed by printed electronics, involve the use of inks or pastes composed by functional materials such as silver, gold or cooper, representing a high intake of precious materials [1]. These metals are used due to their high electrical conductivities (up to 10^7 S/m), while other conductive materials as carbon or PEDOT:PSS (Poly(2,3-dihydrothieno-1,4-dioxin)-poly(styrene sulfonate)), present lower conductivities (up to 10^5 S/m) [2]. Inks and pastes developed for printed electronics are not only composed of these functional materials, but also by polymeric resins or binder, solvents and additives. These materials are not always sustainable, entailing a risk for the environment. In addition, due to the increment on the demand of electronics during the last decades, the electronic waste has increased[3]. On this context, there is a need of developing new inks for printed electronics, based on sustainable, biobased, or biodegradable materials, to reduce the electronic waste and the harm to the environment.

Printed electronics inks or pastes could be used to develop multiple systems or applications. A specific example of an application are sensors. Sensors are electronic designs capable of detecting changes on the environment, by detecting the modification of a physical variable, such as the temperature, velocity or pressure, among others. Sensors detect the change of the physical variable and transform it into an observable electrical response [4]. Currently, they exist commercially a huge number of types of sensors, to detect different stimulus, manufactured using printing technologies. An example of these sensors are temperature sensors, specifically resistive temperature sensors (RTD). The RTDs vary their electrical resistance with the variation of the environment temperature. RTD response is characterized by the Temperature Coefficient Resistance (TCR) of the material the RTD is composed of. The response of an RTD is defined by the equation 1.

$$\alpha = 1 / R(T_0) \bullet (R(T) - R(T_0)) / (T - T_0) \qquad (1)$$

On equation 1, α is the temperature coefficient resistance, T0 is the reference temperature, T is the measuring temperature and R(T) is the resistance of the material at a determined temperature T.

Authors have developed RTD by printing techniques with different materials. Barmpakos et al. [5] designed a temperature sensor onto photographic paper substrate, printed with carbon based and PEDOT:PSS based inks by inkjet printing. Developed sensors could measure temperatures between 25ºC and 45ºC, obtaining positive TCR for carbon printed sensors and negative TCR for PEDOT:PSS printed sensors. Jäger et al. [6] printed resistive temperature sensors by inkjet printing with a silver nanoparticle ink and was characterized between 10ºC and 85ºC at 60% RH. Le Goupil et al. [7] developed screen printed silver-based temperature sensors on different types of substrates obtaining linear electrical resistance variation with temperature, between 20ºC and 40ºC.

Flexible and stretchable devices have gained interest, specifically on wearable technologies [8]. Typical used substrates to give flexibility to sensors are polydimethylsiloxane [9] (PDMS), polyimide [4] (PI), polyethylene terephthalate [10] (PET) based films, polyurethane [11] (PU), papers [12] or fabrics [13], such as glass fiber or carbon fiber.

On this work, temperature sensors will be developed with different pastes for screen printing. Used pastes include commercial silver and carbon pastes and developed carbon pastes with more sustainable materials. The sensors were printed onto PET substrate and onto a glass fiber to consider the possibility of integrating these sensors on composites.

II. MATERIALS AND METHODS

A. *Materials*

On this work, two different pastes were prepared.

1) Paste A

For the synthesis of Paste A, multiwall carbon nanotubes (MWCNT) were used as functional material. N-butyl acetate was used as solvent. For the polymeric binder of the paste, a natural origin epoxy resin was used, supplied by Traquisa (Barcelona, Spain). As curing agent, vanillin-derived Schiff base was synthesized and used (Monteserin et al. [14]).

2) Paste B

For the synthesis of Paste B, porous carbon nano powder (plant based) was used as conductive material. Carbon particles were dispersed on a PEDOT:Biopolymer water dispersion provided by POLYMAT (University of the Basque Country (UPV/EHU)) [15]. As binder, gelatine coming from fish source was used.

B. *Pastes synthesis*

For both pastes, functional particles were dispersed on its solvent by ultrasound (US) sonication. Afterwards, the polymeric binder was added, and the pastes were mixed by mechanical stirring until they get paste appearance.

C. *Sensor development*

On this work, temperature sensors were developed, printing them by screen printing onto glass fiber substrate. The pastes used to print those sensors were the two developed pastes (Paste A and Paste B), a commercial silver-based paste and a commercial carbon-based paste. After printing, the deposited pastes were cured using a box oven. The sensors were infused in an epoxy resin to protect them.

D. *Characterization methods*

1) Viscosity measurements

Screen printing pastes must have no Newtonian behavior. This behavior was characterized by using a rheometer and measuring the viscosity of the pastes modifying the applied shear rate from 0,1 per second to 1000 per second.

2) Conductivity measurements

Sheet resistance measurements were performed using the four-point probe on a deposited screen-printed layer. For measuring conductivity, the thickness of the layer was measured.

3) Temperature sensor behavior

To characterize the sensors behavior, a climatic box was used. The climatic conditions were modified between 15ºC and 50ºC at a humidity of the 40% RH, and sensor resistance was measured each 10 seconds, to analyze the variation of resistance with the applied temperature.

III. RESULTS

A. *Conductive paste synthesis results*

By dispersing carbonaceous nanoparticles on the selected solvents and mixed with the polymeric binder, in each case, conductive pastes were achieved after performing mechanical stirring.

Viscosity measurements were performed for commercial and prepared pastes, and no Newtonian behavior was confirmed, as it is shown in Fig. 1.

Conductivity of each of the samples was confirmed, achieving a conductivity of 6,2 siemens per meter for paste A, 4 siemens per meter for paste B, $8 \cdot 10^5$ siemens per meter for silver commercial paste and 350 siemens per meter for carbon conductive paste. As it is shown, the conductivities of the synthesized pastes are below the conductivity of the commercial carbon paste, which could lead into a better behavior to use them as specific sensors.

After having measured paste viscosities, resistive temperature sensors were printed by screen printing, and cured in box oven. The sensor was a coil design, and its traces widths were adjusted to ensure conductivity onto glass fiber substrates. By printing these sensors, the printability of the synthesized pastes by screen printing onto glass fiber substrate was demonstrated.

Once the printed design was cured, the sensors were infused with an epoxy resin and its hardening agent to protect them from humidity. The mix of epoxy resin and hardening agent was deposited onto the glass fiber with the printed sensor and pressed between two glasses, to ensure all the sensor surface is covered with the mix. The system was dried for 1 day at room temperature. After this time, pressing glasses were removed and the sensor was embedded in the epoxy resin. Printed sensors are shown in Fig. 2.

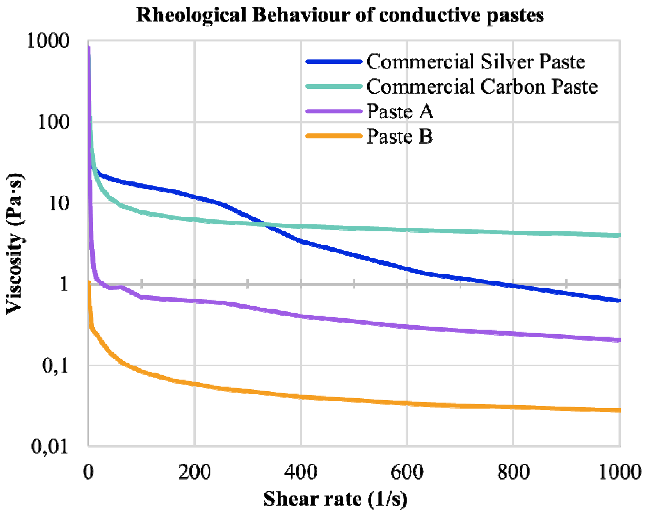

Fig. 1. Rheological behavior of screen-printing pastes

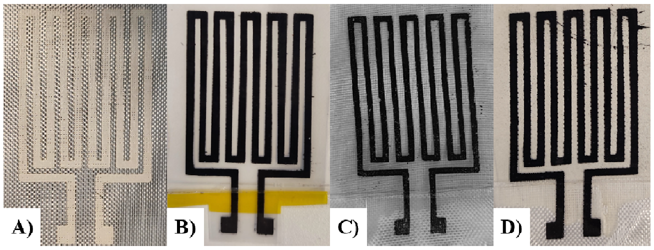

Fig. 2. Printed temperature sensors: A) Commercial silver paste, B) Commercial carbon paste, C) Paste A and D) Paste B.

After the sensors were protected, they were characterized on a climatic box, at a humidity of 40% RH, from 15ºC to 50ºC. The reference temperature was taken at 15ºC.

The obtained tendencies are shown on Fig. 3 As it is shown, silver sensor presents a linear tendency, obtaining a temperature coefficient resistance of 0,003 per centigrade, which is comparable with literature ($3.8 \cdot 10^{-3}$ per centigrade [16]).

On the other hand, carbon paste sensors present different behaviors depending on the carbonaceous materials. These behavior was shown by Wu et al. [17] who developed different temperature sensors with graphitic flakes and carbon nanotubes, obtaining opposite temperature coefficient resistance values.

Commercial carbon-based paste exhibits a polynomic behavior (grade 2). Nevertheless, as the second-grade contribution is really small, a lineal approximation could be done obtaining a positive temperature coefficient resistance of 0,0015 per centigrade.

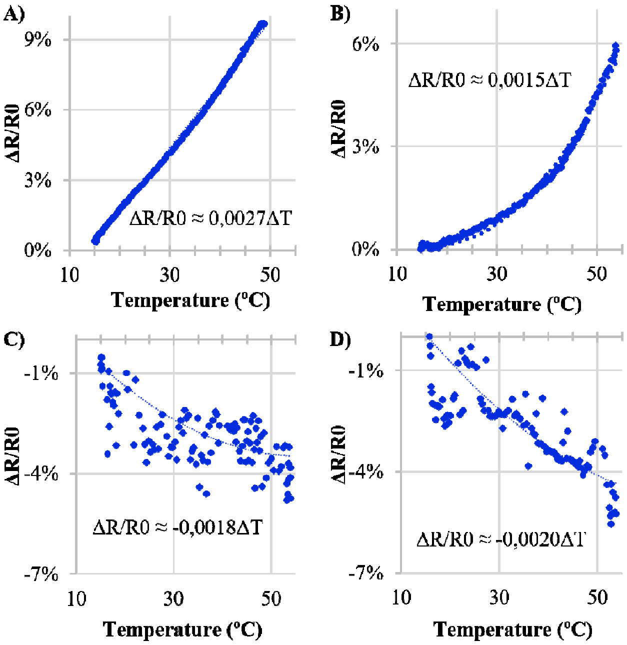

Fig. 3. Electrical resistance of the different material screen-printed sensors: A) Commercial silver based paste sensor behavior, B) Commercial carbon based pastes sensors behavior, C) Paste A sensor behavior and D) Paste B sensor behavior.

Prepared pastes also present a second-grade polynomic behaviour, and the same linear approximation could be made. In both cases, the obtained temperature coefficient resistance is negative value, achieving -0.0018 per centigrade and -0.0020 per centigrade for pastes A and B respectively.

For carbon made pastes, the obtained temperature coefficient resistance is higher than the carbon by itself found in literature, being this $-5 \cdot 10^{-4}$ per centigrade [16]. The difference on the tendencies (positive or negative) could be explained as the contribution to the temperature coefficient resistance of the matrix the particles are dispersed in, changing, in the commercial paste, the value from negative to positive. The increase in the temperature coefficient resistance could be explained by the combination of the varied materials that conform the paste, which could be altering this value.

On Fig. 3 could be noticed that for prepared pastes the data dispersion is not as accurate as the commercial pastes data. These denote that these manufactured sensors could be improved by modifying the composition and preparation method of the pastes or by changing the printed design.

IV. CONCLUSIONS

The need to develop more sustainable electronic systems is a reality on recent times. This goal could be reached by using more sustainable or biobased materials as well as avoiding environmental harmful materials and products.

On this work, more sustainable pastes have been developed for screen printing, and their printability has been demonstrated. The pastes have been used to develop printed temperature sensors deposited onto glass fiber and protected with an epoxy resin. Their use as temperature sensors has been demonstrated by measuring their resistance variation with temperature and their obtained temperature coefficient resistance has been compared with the theorical ones. Nevertheless, better combination of materials and sample preparing processes may be performed to ensure a better behavior.

Due to the selected printed substrate and the flexibility of the final sensors, in combination with the exhibited behavior, these sensors could be a good option to be used as wearable devices.

ACKNOWLEDGMENT

This publication is supported by the SUINK project funded by the European Union's Horizon Europe research and innovation program under Grant Agreement No. 101070112.Views and opinions expressed are, however, those of the author(s) only and do not necessarily reflect those of the European Union or the European Commission. Neither the European Union nor the granting authority can be held responsible for them.

The authors would like to acknowledge the Basque Government funding within the ELKARTEK2023 KK-2023/00056 Programs.

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