DOI 10.5162/IMCS2012/P1.9.12

Fully printed wireless LC sensor for heavy metal detection

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Abstract:

This paper reports on the successful development of a fully printed wireless LC sensor for the detection of toxic heavy metals. The sensor, consisting of an inductor, detection coil and interdigitated electrodes (IDE) in planar form, was fabricated using screen and gravure printing technologies on a flexible polyethylene-terephthalate (PET) substrate with silver based ink as metallization. The capability of the printed LC sensor for detecting very low concentrations of toxic heavy metals was demonstrated. The wireless response of the printed LC sensor revealed a very high sensitivity at picomolar levels of cadmium sulphide (CdS) and lead sulphide (PbS).

Key words: Printed electronics, LC sensor, wireless, gravure printing, screen printing, heavy metals.

Introduction

Over the past decade, a lot of interest has been centered on the development of efficient, low cost and portable sensing systems for the detection of heavy metals in the food processing and environmental industries [1-4]. Toxic heavy metals, which are major environmental pollutants in land and water, are non-biodegradable and therefore remain in the ecosystem and food chain [5]. They have a high potential of causing various disease in humans and animals like renal failure, chronic toxicity, liver damage and ultimately loss of life [6]. Studies have reported on the use of various techniques for the detection of toxic heavy metals. To name a few, titration method [7], colorimetric analysis [8] and electrochemical impedance spectroscopy [9]. Although these techniques are sensitive enough, they require complicated instrumentation along with high manufacturing and operational costs.

In recent years, research has been focusing towards the development of low cost wireless sensors for applications in the environmental, food and biomedical industries [10-11]. Wireless sensing is usually performed in active and passive sensing methods. Active sensing method uses amplifiers, radio frequency (RF) circuits, batteries and antennas which make the sensors complex and expensive [12]. On the other hand, passive sensing method uses a basic inductive capacitor (LC) circuit that does not require complex circuits, are battery free and most importantly cost effective when compared to the active wireless sensors [13]. LC sensors are usually fabricated using conventional CMOS based techniques, which are typically expensive. The recent trend of using traditional printing methods in the field of printed electronics (PE) is a promising technique for the fabrication of low cost sensors.

PE is emerging as an alternate solution to traditional CMOS based techniques for fabrication of electronic devices which requires complex fabrication steps, needs high operating temperatures and ultimately is a costly process [14]. PE uses additive printing methods that can work on flexible substrates, uses less complex fabrication steps and reduces wastage of resources which leads to a cost effective fabrication method. Among all the printing methods, gravure, screen, inkjet and flexographic printing techniques have received significant attention for the fabrication of electronic devices such as displays [15], thin film transistors (TFT's) [16] and solar cells [17]. Although, recent studies have been reported on the fabrication of sensors using conventional printing techniques, there are no reports on the printing of wireless flexible LC sensors for toxic heavy metals detection.

In this work, the gravure and screen printing techniques was used to fabricate a fully printed LC sensor on a flexible substrate using silver based inks as metallization. Quantitative

detection of toxic heavy metals like cadmium sulfide (CdS) and lead sulfide (PbS), was used to demonstrate the feasibility of the printed LC sensor.

Screen printing

Screen printing is a push through process in which the substrate is not in direct contact with the mask or image carrier (as shown in Fig. 1). The main components of the screen printing are a squeegee and a screen printing plate. The ink is applied on top of the screen over which the rubber squeegee is swept over with high pressure. This causes the ink to pass through the screen and is transferred onto the substrate [18].

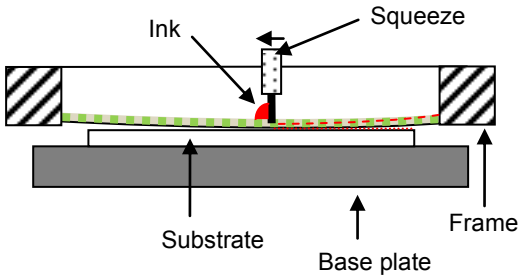

Fig. 1. Screen printing.

Gravure printing

Gravure printing is known for its high quality printing, fast output, robustness of the process and use of low viscosity inks. Image carrier (gravure cylinder), ink fountain, doctor blade and impression cylinder are the basic components of a typical gravure system (as shown in Fig. 2). The gravure cylinder has small cells or image areas on the surface that are responsible for carrying ink from the ink fountain to the substrate. Transferring the ink from the cells onto the substrate is assisted by the impression cylinder [19].

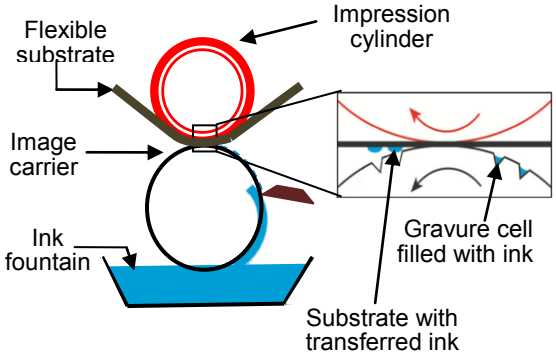

Fig. 2. Gravure printing.

Sensor fabrication

The passive LC sensor was fabricated using the gravure and screen printing techniques. Melinex ST 506, flexible polyethylene-terephthalate (PET) from DuPont Teijin Films was used as a substrate. TEC-PR-020, a silver nanoparticle based ink, and Electrodag 479SS, a silver ink, from Henkel was used for metallization in gravure and screen printing, respectively.

The wireless LC sensor consists of printed interdigitated electrodes (IDE's) and an inductor in planar form. Fig. 3(a) shows the screen printed flexible planar inductor with electrode dimensions of 200 µm line width and 12 turns. The gravure printed IDE's, consisting of 8 pair of electrodes with 8600 µm length, 200 µm width and spacing, is shown in Fig. 3(b). The printed inductor was attached to the printed IDE's using connecting wires and Ag conductive epoxy (MG chemicals®) to form a resonant LC circuit.

Experimental

CdS and PbS (in crystalline form) were purchased from Sigma–Aldrich Chemical Company. CdS and PbS were dissolved in deionized water (DI) to obtain concentrations of 1 pM, 100 pM, 1 nM, 100 nM and 100 µM. Fig. 4 shows the block diagram of the experiment setup. An Agilent impedance analyzer (4395A) was used to remotely monitor the LC sensors by measuring the impedance of the detection coil (screen printed planar inductor). Calibration for wires and probes was done before taking measurements. The data was recorded and analyzed using a custom built LabVIEW program on PC connected to the impedance analyzer via GPIB cable. 100 µl of varying concentrations of CdS and PbS were loaded onto the LC sensor using a pipette. The frequency spectrum, from 32 MHz to 33 MHz,

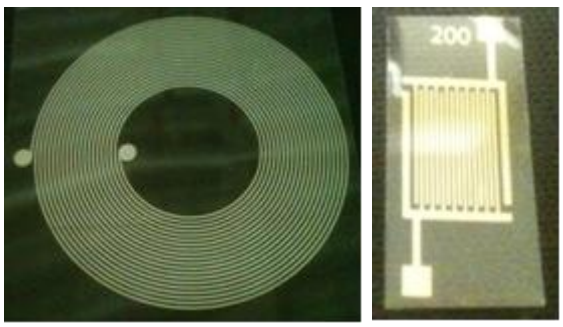

Fig. 3. (a) Screen printed planar inductor and (b) Gravure printed IDE's.

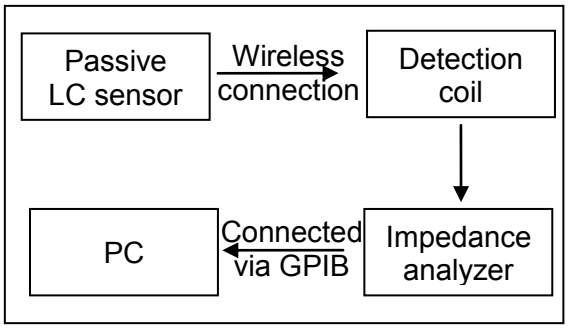

Fig. 4. Experiment setup.

of the printed LC sensor was wirelessly monitored by the detection coil. All the measurements were conducted at room temperature.

Results and Discussion

The response of the printed LC sensor was first tested towards CdS. A reference signal was established by placing 100 µl DI on the IDE's. The phase shift of the LC sensor was monitored for various concentrations of the CdS. The change in frequency of the LC sensor with respect to concentration of CdS is shown in Fig. 5. The resonant frequency change (Δf) of the LC sensor was 1.2 kHz, 2.5 kHz, 16.2 kHz and 40 kHz when compared to base DI, for 1 pM, 100 pM, 1 nM, and 100 nM concentrations of CdS, respectively. It is worth noting that the detection of CdS was made possible at concentrations as low as 1 pM while the US Food and Drug Administration (USFDA) toxicity limit of cadmium is 3 µM [20].

Similarly, the response of the printed LC sensor was tested towards varying concentrations of PbS. Fig. 6 depicts the percentage change in resonant frequency of the LC sensor with respect to the change in concentration of PbS. It was observed that the percentage change in frequency was 0.1 %, 0.3 %, 0.35 % and 0.4 % for 1 pM, 100 pM, 100 nM and 100 µM concentrations of PbS, respectively when

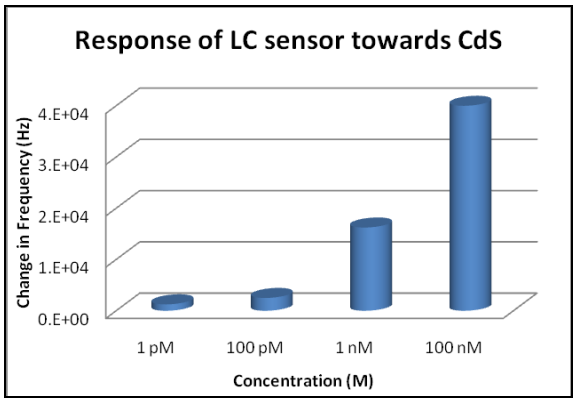

Fig. 5. Wireless resonant frequency response of the LC sensor towards varying concentrations of CdS.

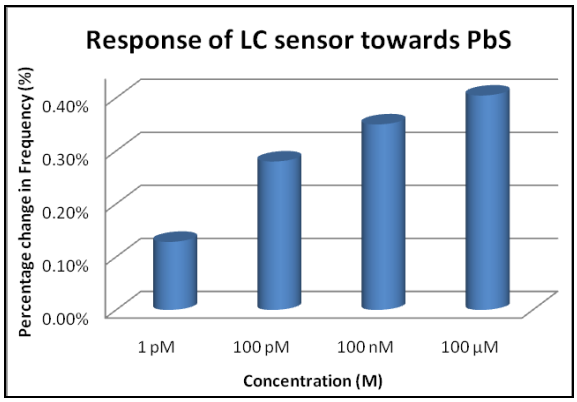

Fig. 6 Percentage change in resonant frequency of the LC sensor towards varying concentrations of PbS.

compared to base DI. PbS was detected at concentrations as low as 1 pM. This is well below the limit of 3 µM regulated by the USFDA [20].

Conclusion

A fully printed LC sensor was successfully fabricated using gravure and screen printing techniques on flexible PET substrate with electrode dimensions of 200 µm width and spacing. Ag based ink was used for metallization. The feasibility of the device to distinguish among various concentrations of heavy metals such as CdS and PbS was demonstrated. The measured change in resonant frequency of the LC sensor towards 1 pM concentration of CdS and PbS showed a variation of 1.2 kHz and 40 kHz, respectively from the base DI. The results obtained show a promising potential of fully printed flexible wireless sensors for the detection of toxic heavy metals. Further research is underway to improve the sensitivity and selectivity of the sensor towards various heavy metal compounds.

Acknowledgement

This work has been partially supported by the U.S. Army Grant 204 no.WS911QY-07-1-0003 and W911NF-09-C-0135.

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