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Radio-frequency tag with optoelectronic interface for distributed wireless chemical and biological sensor applications

Ivana Murković Steinberg[a,*], Matthew D. Steinberg[b]

[a] Faculty of Chemical Engineering & Technology, University of Zagreb, Marulićev trg 19, HR-10000 Zagreb, Croatia
[b] Erasmus Technology LLP, 57A Moorfield Road, Duxford, Cambridge, CB22 4PP, UK

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ABSTRACT

A battery-free, wireless optical chemical sensor with integral contactless power and data link is demonstrated. The chemical sensor and its optoelectronic interface form an integral part of a radio-frequency tag which we have developed specifically for use as a wireless chemical and biological sensor, and which is compatible with the International Standards Organisation ISO15693 radio-frequency identification (RFID) protocol. The chemical sensor, optoelectronic interface, RFID processor and antenna are designed and fabricated on a credit-card sized tag which is operated in a passive mode using wireless energy transfer to power the sensor and read its response. The optical chemical sensing capability of the RFID tag is demonstrated by measuring pH using a thin sol–gel film containing a pH-sensitive indicator dye, bromocresol green (BCG), as a model chemical sensor interface. The film exhibits a colourimetric response to pH in the range 5.0–8.5 pH units. The optoelectronic interface of the tag comprises a silicon photodiode and two light emitting diode (LED) sources measuring the optical absorption of the pH-induced colour change of the thin film at two discrete wavelengths. The pH response curve and the p*K* value of the immobilised indicator were determined using the RFID tag system and compared with the results obtained using a standard laboratory spectrophotometer. A good correlation between the measurements was achieved, demonstrating the viable integration of an optical absorption based chemical sensor onto a passive RFID tag. By combining the relative advantages of simple, tuneable, optical sol–gel sensing chemistries and integrating these onto the RFID wireless communication platform, it is possible to envisage in the near future a range of new wireless sensors for distributed industrial, scientific and medical applications, in particular for smart packaging, personal (wearable) diagnostics, and infrastructure-related applications.

1. Introduction

The mobile and wireless communications revolution is entering a new phase as it stimulates markets for ubiquitous, pervasive, and wireless data applications, including sensor networks [1]. This new phase is driven by declining cost of ownership and increasing maturity and adoption of wireless standards, such as Bluetooth, ZigBee, WiFi and radio-frequency identification (RFID) which allows global interoperability between devices and device manufacturers.

The RFID protocol was originally developed for short-range product identification, typically covering the 2 mm–2 m read range, and has been promoted as the replacement technology for the optical bar-code found on fast moving consumer goods. However, growing interest in the many possible applications of wireless chemical sensors has led to the development of a number of RFID-based physical and chemical sensors. As reported in the literature, these range from the relatively straightforward measurement of temperature of perishable foods [2], to more sophisticated multi-gas detection with on-tag metal-oxide gas sensors [3], to mixed-vapour detection with polymer electrolyte membranes (PEMs) together with more complex, off-tag, signal processing techniques [4]. The fusion of chemical and biosensor technologies with RFID and the other short-range, low-cost, and ubiquitous wireless technologies is creating new opportunities for sensing applications in healthcare, environmental and building infrastructure monitoring, process and quality control, and chemical and biological threat detection [5]. The ability to gather information from spatially distributed sensors with the innate ability to communicate amongst themselves, and to form and configure different network topologies is opening a diverse range of new applications in sensor science [6,7]. Wireless sensor networks (WSNs) provide the technological infrastructure for realising such distributed systems, and the underlying topological theories, enabling hardware and software technologies, and diverse applications of WSNs have been widely reported [8–10].

* Corresponding author.
E-mail addresses: ivana.murkovic@fkit.hr (I.M. Steinberg), matthew.steinberg@erasmus.eu.com (M.D. Steinberg).

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In addition to wireless RFID-based sensors, other types of passive, contactless chemical sensor have been reported that encompass a wide range of different transduction methods, including surface acoustic wave (SAW) devices [11–14], magnetoelastic devices [15–17], and tuned resonant electric circuit devices based on variable capacitor–inductor (LC) resonant tuned circuits. The LC tags resonate when exposed to an applied radio-frequency (RF) field, and the resonant frequency varies with analyte concentration, either because of interaction with a chemically sensitive dielectric material [18], or because of a conformational change in the gap separating the capacitive plates [19]. We previously reported a passive multi-analyte integrated circuit electrochemical sensor with a RF interface that was developed for use as an *in vivo* implant which made use of a proprietary data transmission protocol based on pulse width modulation (PWM) [20], and another low-cost micropower amperometric potentiostat for contactless operation used a pulse frequency modulation (FM) technique for communication [21]. Both these contactless devices were based on the RFID operating principle, where antenna load-modulation is used to send data from the sensor to a nearby reader, however, neither of these devices could be read with standard RFID readers because of their proprietary data coding schemes.

Such devices demonstrate the feasibility and growing commercial demand for low cost contactless chemical sensors. However, in the majority of cases, these devices do not generate machine-readable digital output data, and often require complex signal processing equipment to deconvolve the analytical signal. So whilst the sensors are often very low-cost, the supporting instrumentation required to read them is not. Most importantly in relation to WSNs, such contactless sensors are not capable of forming or joining networks, and multiple devices may not be queried simultaneously.

In order to address the growing need for contactless chemical sensors with a standardised and machine-readable output format that includes an anti-collision protocol, we are developing tags based on the ISO 15693 radio-frequency identification specification. The work presented here describes an optical chemical sensor (optrode) developed specifically for incorporation onto the passive, battery-free RFID platform.

The chemical interface of the RFID optrode is designed for the measurement of pH using a thin sol–gel film containing the pH-sensitive indicator dye, bromocresol green (BCG). Sol–gel technology is especially convenient for development of integrated optoelectronic sensing devices since it allows for low-temperature preparation of optically transparent porous thin films. Such films have been widely used as solid matrices for immobilisation of chemical and biochemical reagents for optical chemical sensors and biosensors [22], including various innovative applications such as smart food packaging [23]. Because sol–gel thin films may be produced from a single coating cocktail of chemicals, they are both low-cost and suitable for high volume manufacturing, making them viable candidate materials for distributed wireless chemical sensors.

2. Experimental

2.1. Radio-frequency tag

A wireless tag, or vicinity integrated circuit card (VICC), based on the ISO15693 RFID standard has been developed around a commercial microcontroller (PIC12F683, Microchip Technology Inc., Chandler, AZ, USA), and is approximately the size of a credit-card. The microcontroller is connected to a proprietary optoelectronic interface, comprising a surface mount photodiode (BPW34S, Osram Opto Semiconductors GmbH, Regensburg, Germany) and two surface mount light emitting diodes (LED) (HSMx-A10x, Agilent Technologies Inc., Santa Clara, CA, USA) arranged in a planar configuration.

2.2. Materials and methods

Electronic circuits for the wireless tag (vicinity integrated circuit card) were designed using a schematic capture design and printed circuit board (PCB) layout suite (EASY-PC Professional v11.0, Number One Systems Ltd., Gloucester, UK). Printed circuit boards for the VICC and for the optoelectronic interfaces were fabricated from standard FR4 materials (RAK Printed Circuits Ltd., Saffron Walden, UK). Code for the VICC microprocessor was written, developed and programmed into the device's flash memory using an integrated software design environment and device programmer (MPLAB v8.0 with PICStart plus, Microchip Technology Inc., Chandler, AZ, USA). The sol–gel precursor tetraethoxysilane (TEOS) and cetyltrimethyl ammonium chloride (CTAC) were used as supplied (Sigma–Aldrich, BioVit d.o.o., Varazdin, Croatia), BCG indicator was used as supplied (Kemika d.o.o., Zagreb, Croatia). Standard buffer solutions were prepared covering the following pH ranges: citrate buffer (pH range1 to 4), acetate buffer (pH range 4–5) and phosphate buffer (pH range 5–9).

2.3. Optrode interface

pH sensitive sol–gel thin films were prepared from a cocktail as previously reported [24]. Briefly, 1 mL of sol–gel precursor TEOS, 1.2 mL of ethanol, 0.4 mL of 0.1 M HCl, 16.8 mg of BCG indicator and 0.2 mL of CTAC solution (2×10^{-3} M) were mixed and homogenised for 10 min in an ultrasonic bath (Transsonic T 460/H, Elma Hans Schmidbauer GmbH & Co. KG, Singen, Germany), and the sol–gel solution then allowed to gelate at room temperature for 48 h. Glass substrates measuring 2.5 cm × 2.5 cm × 1.0 mm were cut from standard microscope slides, and treated with concentrated nitric acid for 12 h in order to activate the silanol groups on the surface of the glass. Substrates were rinsed with ethanol and distilled water and dried at 100 °C for 3 h. Optical were obtained by spin-coating 100 μL of the gelated cocktail at 3500 rpm for 30 s onto the substrates (P-6204 spin coater, Speedline Technologies Inc., Franklin, MA, USA). After spin-coating, films were dried at room temperature and stored in the dark for 72 h. Smaller glass chips with dimensions of 1 cm × 2.5 cm were cut from the coated substrates in order to perform the spectral characterisation. Glass chips were dipped in buffer and either placed directly onto the optoelectronic interface of the VICC tag, or were placed along the wall of the sample cuvettes of a laboratory spectrophotometer and exposed serially to the different buffer solutions. The measurement procedure was repeated with different buffers covering the pH range from 3.8 to 9.0 pH units. Absorption spectra were measured with the UV–vis spectrophotometer (DMS 80, Varian Inc., Palo Alto, CA, USA) and with the VICC tag. Homogeneity of the films was visually inspected using an optical stereo microscope (BX 50, Olympus Corp., Tokyo, Japan).

3. Results

3.1. Wireless sensor platform

The wireless sensor platform comprises a commercial RFID reader connected to a personal computer, Fig. 1. The RFID reader energises and communicates with passive tags (VICCs) over a range of up to 2 m away. Passive tags do not contain a battery or power supply, but operate by scavenging energy from the electromagnetic field generated by the antenna connected to the reader. The main components of the chemical sensor VICC are the antenna, the RFID