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Multimodal technique to eliminate humidity interference for specific detection of ethanol



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ABSTRACT

Multimodal electrochemical technique incorporating both open circuit potential (OCP) and amperometric techniques have been conceptualized and implemented to improve the detection of specific analyte in systems where more than one analyte is present. This approach has been demonstrated through the detection of ethanol while eliminating the contribution of water in a micro fuel cell sensor system. The sensor was interfaced with LMP91000 potentiostat, controlled through MSP430F5529LP microcontroller to implement an auto-calibration algorithm tailored to improve the detection of alcohol. The sensor was designed and fabricated as a three electrode system with Nafion as a proton exchange membrane (PEM). The electrochemical signal of the interfering phase (water) was eliminated by implementing the multimodal electrochemical detection technique. The results were validated by comparing sensor and potentiostat performances with a commercial sensor and potentiostat respectively. The results suggest that such a sensing system can detect ethanol at concentrations as low as 5 ppm. The structure and properties such as low detection limit, selectivity and miniaturized size enables potential application of this device in wearable transdermal alcohol measurements.

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1. Introduction

Continuous wearable alcohol measurement system has been sought in numerous fields ranging from law enforcement to clinical monitoring to safety systems (Goodman et al., 1986; Highway Traffic Safety Administration and Department of Transportation, 2013; Room et al., 2005; WHO, 2015). Breath alcohol (BA) measurement devices are used by law enforcement agencies for the random monitoring of drivers to determine whether they are driving under influence of alcohol (Mason and Dubowski, 1974; Dubowski, 1994; Zuba, 2008; Polissar et al., 2015). Since law enforcement involves measuring the BA content of a random population at one point in time, breath analyzers are adequate. However, in a clinical application focused on understanding consumption and metabolism of alcohol, measurements are required over extended periods, ideally starting from 30 min before consumption and extending to 8 h after the last drink. Given that alcohol is generally consumed in the evening and the subject will likely sleep following his ingestion of alcohol, a continuous alcohol sensor is preferred. The simplest approach to continuous monitoring of alcohol is a wearable sensor that uses alcohol vapors emanating from the skin and secreted with sweat (Sakai et al., 2006).

After alcohol ingestion, a very small proportion (1% of overall alcohol consumption) is excreted with sweat from the exocrine sweat gland or in a diffusive manner (Marques and McKnight, 2009). The sweat is mostly composed of water (\sim 99% of overall volume) with small amount of nitrogenous compounds, metal and nonmetal ions, metabolites, xenobiotics, organic volatile compounds and so on (Jadoon et al., 2015). Hence, elimination of cross selectivity with different compounds, especially with the water content of sweat or atmospheric humidity is challenging for any transdermal alcohol sensors. The reaction kinetics (Zhang et al., 2008) and maximum power density (Lee and Hwang, 2009) of the fuel cell sensor are directly dependent on relative humidity. In the electrochemical sensing, therefore, humidity has an adverse impact on the calibration of the sensors. Similar to humidity, other volatile compounds released from the skin such as aldehydes and ketones (Mochalski et al., 2014), will undergo reaction in the anode creating interference signal. Only a selective and accurate alcohol measurement device would be the possible solution to overcome this issue. This can be achieved in two ways: firstly, to develop a highly selective sensor that can afford accurate signal for

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only alcohol or secondly, electrically eliminate the noise signals following a robust electrochemical technique.

Current alcohol sensors in the market use five measurement techniques for monitoring alcohol: optical (Sanford et al., 2001; Shabaneh et al., 2014; Semwal et al., 2016), colorimetric (Williams and Reese, 1950; Lidén et al., 1998), electrochemical (Santra et al., 2016; Jiang et al., 2016; Shan et al., 2010; Kim et al., 2000; Swift et al., 1992), biomarker sensing (An et al., 2014) or solid-state techniques (Yu et al., 2012; Kadir et al., 2014; Kim and Lee., 2014). Most of these sensors suffer from instability, non-linearity, cross-selectivity, inaccuracy at low concentrations, and portability (Paixão and Bertotti, 2004). Electrochemical alcohol sensors have typically been found to be most suitable for long term sensing. Among them, fuel cell sensors offer simplicity, relatively high accuracy, sensitivity, long working lifetime, scalability and portability (Marques and McKnight, 2009), hence they find use in breathalyzers.

Wearable transdermal alcohol measurement devices have been built before e.g. 'GinerWrisTAS' (Injury Research Foundation, 2007). However, this device did not have a data acquisition system and required a humid chamber for measurements. Others (Injury Research Foundation, 2007) integrated the data acquisition system and incorporated PEM as an electrolyte. These sensors suffered from false positive readings due to the presence of volatile organic compounds, lack of selectivity, and needs for frequent manual calibrations. Both fuel cell/electrochemical breathalyzers and wearable devices measure interactions between the sensor and volatile organic compounds (VOCs) such as acetone, acetophenone, isoprene, etc (Mochalski et al., 2014; Acevedo et al., 2007). The interactions lead to change in potential and generate a signal. When these devices are used to for measurement of alcohol, the VOCs trigger false positives.

To address the needs of the target users, a complete wearable system needs to be developed. Such a system should be able to recalibrate and reference itself to its environment, minimizing the effect of VOCs, thus enabling long term quantitative monitoring. The platform needs to provide real-time statistical analysis for different vapor footprints, including alcohol so it can be calibrated to the needs of the user. Such systems should also be low powered to ensure long usage life between charges. This paper presents such a sensing system that has the ability to communicate with external devices through Bluetooth for longitudinal data analysis to ensure functionality. Finally, the sensor was packaged into a wristwatch format.

2. Experimental

2.1. Materials and methods

Perfluorinated Nafion424 reinforced with poly-tetra-fluoroethylene (PTFE) fibers (thickness 0.03 cm) as a PEM, nickel chloride anhydrous, nickel sulfamate, boric acid, 95% sulfuric acid (H₂SO₄) and 37% hydrochloric acid (HCl) were purchased from Sigma-Aldrich. Lead and nickel sheets were purchased from McMaster-Carr for the electroplating process. Acetone and ethanol (95.27%) were purchased from Fisher scientific Inc. All other used chemicals were of analytical grade. The preparation of aqueous solutions was done with de-ionized (DI) water.

MICRO5 PID sensor purchased from BW Technologies. Potentiostat (CHI 1230B with MC470) purchased from CH instruments Inc. LMP91000 miniaturized potentiostat with analog front end (AFE), a 16-bit ultra-low power microcontroller (MSP430F5529LP), RN42 Bluetooth chip and LP2591 power management system were purchased from Texas Instruments (TI). MCP72831 charge controller, 12 bit digital-to-analog converter

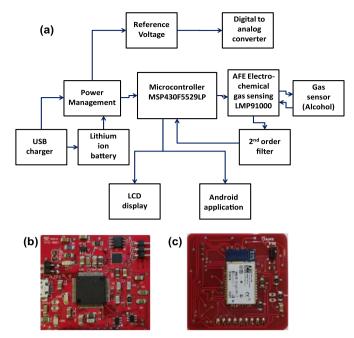


Fig. 1. (a) Block diagram and schematic of peripheral connections in the alcohol monitoring device, (b) top and (c) bottom layer of the PCB board where the top layer consists of LMP91000, microprocessor and other integrated electronic components and the bottom layer consist of Bluetooth component.

with integrated electrically erasable programmable read-only memory (EEPROM) and an I2C compatible serial interface from Microchip were purchased.

2.2. Potentiostat platform and fuel cell sensor construction

The fuel cell sensor was interfaced with the AFE (LMP91000) on a wearable platform. The working electrode (WE), reference electrode (RE), and counter electrode (CE) of the sensor were connected to the corresponding pins of the AFE. The AFE was linked to the microcontroller via inter-integrated circuit (I2C) interface. The Bluetooth module was connected via universal asynchronous receiver/transmitter (UART) peripheral of the MSP430. The liquid crystal display (LCD) used the serial peripheral interface (SPI) of the MSP430. The connections and their functions are discussed in Section 3. The peripheral connection is depicted in the block diagram as shown in Fig. 1a.

The printed circuit board (PCB) was manufactured on a two-layer board using a 1.6 mm thick FR-4 substrate with 28.35 g copper and an area of 6.45 cm². All the electronic components including the microcontroller, AFE, power management system, and associated circuitry were mounted on the top layer as shown in Fig. 1b. The Bluetooth chip was mounted on the bottom layer Bluetooth solely (Fig. 1c).

Plated 200 μm thick micro-perforated stainless steel sheet with 180 μm pore was used to form WE and CE. A non-conventional standardized material was used for RE. Nafion was hot-pressed between the electrodes to form the fuel cell. The catalyst (nickel) coating on the micro-perforated stainless steel sheet electrodes was achieved by a five step process: (i) anodic cleaning of stainless sheet for 5 min with 25% H_2SO_4 below room temperature at a current density of 13.94 A m $^{-2}$. In electro-cleaning setup, the stainless steel served as the anode and the lead sheet served as the cathode; (ii) acid cleaning the stainless sheets at room temperature with 1:10:1000 solution of HCl, H_2SO_4 and DI water; (iii) Wood's nickel strike using 1.5 M HCl and 1.009 M anhydrous nickel chloride solution and by applying current densities of 4.65 A m $^{-2}$ and 1.4 A m $^{-2}$ for 2 min each consecutively (Harding and Di Bari,

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