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Wearable Autonomous Microsystem with Electrochemical Gas Sensor Array for Real-Time Health and Safety Monitoring

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Abstract—**Airborne pollution and explosive gases threaten human health and occupational safety, therefore generating high demand for a wearable autonomous multi-analyte gas sensor system for real-time environmental monitoring. This paper presents a system level solution through synergistic integration of sensors, electronics, and data analysis algorithms. Electrochemical sensors featuring ionic liquids were chosen to provide low-power room-temperature operation, rapid response, high sensitivity, good selectivity, and a long operating life with low maintenance. The system utilizes a multi-mode electrochemical instrumentation circuit that combines all signal condition functions within a single microelectronics chip to minimize system cost, size and power consumption. Embedded sensor array signal processing algorithms enable gas classification and concentration estimation within a real-world mixture of analytes. System design and integration methodologies are described, and preliminary results are shown for a first generation SO_2 sensor and a thumb-drive sized prototype system.**

I. Introduction

Exposure to air pollution consistently ranks among the leading global causes of illness and mortality [1], and explosive gases are an increasing threat to occupational safety as energy demands rise. Airborne pollutants and explosive gases vary in both time and space. For example, CO and CH_4 can be released from boilers and stoves in homes, and dangerous levels of CH_4 and SO_2 can be found in underground coal mines [2]. To improve scientific understanding of the health impacts of personal exposure to these pollutants, a miniaturized monitoring device is needed that individuals can wear or carry to constantly examine their surrounding environment. To be the most effective, the monitoring device must be cost-effective, allow real-time data collection, and operate autonomously with no user training or regular maintenance. Toward this goal, significant energy has been devoted to the research and development of gas sensors. For example, an autonomous gas measurement system with infrared gas sensors and wireless transmission has been reported [3], but its handheld size and other restrictions make it unsuitable for long-term individual use. Also, a single-chip gas recognition system has been reported [4], but this device can not measure gas concentrations and lacks many elements of a complete system. Finally, a real-time gas sensor array test system was presented in [5], but it is neither wearable nor autonomous. In summary, although there have been many important advances in gas sensor technologies, development of a wearable, autonomous multi-analyte gas sensor system remains an open challenge.

This paper presents a new system designed to meet the goals for personal exposure assessment and environmental monitoring by integrating sensor arrays, electronics, and data analysis algorithms into an autonomous microsystem. Electrochemical sensors were chosen to provide rapid response, high sensitivity, and good selectivity. The sensors utilize ionic liquid interfaces for low-power room-temperature operation with low maintenance requirements. CMOS instrumentation supporting multiple electrochemical techniques is employed to enable an autonomous measurement system with minimum cost, size and power consumption. Sensor array signal processing algorithms running within the system provide the gas classification and concentration estimation necessary for the mixture of analytes found in real-world environments. Integrating these components into a thumb-drive sized package, the resulting system supports continuous, real-time measurement of hazardous gases within a wearable autonomous platform.

II. Microsystem Design

Simultaneously achieving all of the desired features for a personal exposure monitoring system introduces many challenges. To be wearable, the system needs to be miniaturized, light weight, battery powered and either store data, transmit it wirelessly, or both. A thumb-drive size system with power consumption less than 1mW would be suitable for this goal. To be autonomous the system needs to perform measurements intelligently and without the need of any external equipment, software, etc. To measure target gases in real time, the entire measurement path, from appearance of the analyte to storage of measurement results, must be completed within seconds. This section analyzes these goals and constraints from a system-level perspective and shows how they can be mapped to component-level decisions.

A. Gas sensor technologies

A wearable autonomous multi-gas sensor microsystem for real-time measurement requires gas sensors to be small (chip-scale), low cost, free of maintenance, highly sensitive, highly specific, fast responding, and inherently low power. The sensor choice must also consider the required measurement techniques and instrumentation electronics, which must adhere to the same requirements as the sensor. The three types of gas sensors that are most commonly used are metal oxide gas sensors (e.g. stannic oxide),

TABLE I
COMPARISON OF GAS SENSOR TECHNOLOGIES

Sensor Type	Advantages	Disadvantages
Metal oxide gas sensor: resistance change when gases adsorb or combust with the catalyst material, e.g. Pt or Pd coated on a wire.	Long operation lifetime; reasonable parameter stability; low cost; wide range of gases; little maintenance; 0-100% LEL	Poor selectivity; major dependencies on ambient temperature and humidity; large power consumption; requires oxygen; degrades as it is used; limited measurement range
Non Dispersive Infrared (NDIR): absorption of IR by gases.	Immune to catalyst poisoning; good sensitivity and selectivity, 0-100% LEL	Detects limited number of gases due to fairly narrow bandwidth generally used; more expensive; high maintenance; high power
Electrochemical sensors: redox activity of various analytes.	Good selectivity; low power consumption; low cost; wide dynamic range (0-100% LEL and 0-100% UEL)	High maintenance if liquid electrolytes are used; suffering from interference

LEL = lower explosive limit, UEL=upper explosive limit, % is a volume concentration unit.

non-dispersive infrared (NDIR) sensors, and electrochemical (EC) sensors [6, 7]. A comparison among these sensors is summarized in Table I. Metal oxide sensors have poor selectivity because all reducing gases in the atmosphere are detected on its surface. NDIR sensors show good sensitivity and good selectivity but necessitate a relatively complicated optical system making it expensive and bulky and requiring operation training. Furthermore, they cannot provide real-time monitoring. Compared with the first two sensor types, EC sensors have good selectivity, low power consumption, low cost and wide dynamic range. In addition, they have also proven to be effective for detecting relevant gases such as CH_4, CO, CO_2, NO, NO_2, SO_2, H_2, and O_2 [8-10]. Although traditional EC sensors suffer from limited specificity and interference, these disadvantages can be overcome with a multi-mode EC sensor array using novel materials and structures. Therefore, EC sensors were chosen in our system for multi-gas measurement.

B. EC gas sensor array

EC gas sensors require the use of an electrolyte, which is an ionically conducting medium that transports charge within electrochemical cells, contacts all electrodes effectively, solubilizes the reactants and products for efficient mass transport, and is chemically and physically stable under all conditions of sensor operation. Traditional electrolytes are classified as liquid electrolytes (aqueous and non-aqueous) and solid electrolytes [11, 12]. Liquid electrolytes have high electrical conductivity but suffer from solvent exhaustion and require periodic electrolyte maintenance. The solid electrolytes overcome this problem but suffer from low electrical conductivity at room temperature. Room-temperature ionic liquids (ILs) are novel gas sensing materials that combine the benefits of both solid and liquid electrolytes. Furthermore, the use of ILs as electrolytes can eliminate the need for a membrane to simplify sensor design. Electrochemical oxidation of NH_3, NO_2 and SO_2 and the electrochemical reduction of O_2 have been reported using ionic liquids (ILs), and some of these IL-based gas sensors have shown wide detection limits, high sensitivity and excellent reproducibility [13, 14].

EC sensors can be operated in different modes. The amperometric mode measures the current generated by reaction of an analyte at an electrode at a fixed or variable potential. The electrochemical impedance spectroscopy (EIS) mode measures impedance changes in the double-layer capacitance and the charge-transfer conductance. The sensitivity of an amperometric gas sensor is proportional to its electrochemically active surface and drift over the time. EIS mode can monitor for changes in the active electrode area independent of gas concentrations. Therefore, EIS permits self-monitoring of the sensor's stability and automated calibration for drift mechanisms. EIS also provides an orthogonal detection mode to amperometric mode.

A gas sensor's selectivity is important to a multi-gas measurement system. This characteristic of EC sensors is often based on the value of the working electrode potential, which depends on the electrolyte and working electrode material. Therefore, by optimizing the combination of sensor operating potentials, electrolytic medium (e.g. ILs) and working electrode material, the best transducer interfaces for the IL-based EC sensor array can be achieved for high selectivity of mixed gases. Table II summarizes how the characteristics of the sensor array affect the desired system performance goals.

TABLE II
DESIRED SENSOR PERFORMANCE AND SENSOR ARRAY CHARACTERISTICS

System Performance	Relevant Sensor Characteristics
Sensitive	IL is an exciting material for EC gas sensors and has been show to be very sensitive to the gas analytes at ppb or ppm level detection limits.
Selective	Two dimensional IL amperometric and EIS sensor array with sensor array pattern recognition increases accuracy of detection.
Rapid (real time) response	Both amperometric and EIS sensors permit rapid (near real time) measurement.
Long life time and low maintenance	ILs are chemically stable in normal atmosphere. Their thermostability allows the detection interface to be regenerated by heat.
High reliability	Absence of moving parts and semi-solid state sensor design. Complementary transducer types and can cross verify each other.
Low cost	Amperometric and EIS are low cost and low power transducers readily miniaturized using low-cost batch-fabrication microsystem technologies.
Low power	The transducers require no power and can be measured using low power instrumentation electronics compatible with portable and long-life implementation.

C. *Instrumentation circuits and sensor data processing algorithms*

A sensor array instrumentation circuit is needed to capture and digitize the sensor signal for storage, transmission or further signal processing. The system-level goals place the following requirements on theinstrumentation circuit: low cost, small size, low power consumption, rapid response. Choice of an electrochemical sensor array adds requirements to provide programmable bias potentials (for electrochemical selectivity) and stimulus signals generators supporting amperometric and EIS methods. Our group has prior experience developing CMOS circuits for electrochemical amperometry and EIS [15, 16]. Such circuits show that a highly miniaturized EC sensor array with embedded instrumentation circuitry is feasible.

To maximize the selectivity of an EC gas sensor array, an array signal processing algorithm can be employed. Firstly, a feature extraction method, such as Principal Component Analysis (PCA) or Discriminate Function Analysis (DCA) can be used to extract the features from multiple measurements obtained by the sensor array. Given the extracted features, a classification algorithm can then be applied to classify the presence of certain chemicals or gases. Most of the existing classification algorithms cannot predict concentrations of multiple gases. A new data processing algorithm capable of running within the wearable gas sensor system must be developed to overcome this challenge while maintaining the power budget of the overall system.

III. MCIROSYSTEM IMPLEMENTATION

To achieve all of the design goals outlined in section II, we are developing a compact, multi-analyte, monitoring unit called the intelligent electrochemical gas analysis system, or iEGAS. As shown in Fig. 1, the iEGAS system combines a sensor board and a microcontroller board. The sensor board includes an EC sensor array and its instrumentation circuit. A multi-mode EC instrumentation chip (MEIC) provides stimulus signals for EC sensor array and measures sensor response. Temperature and humidity sensors are included to compensate for secondary sensitivities of the gas sensor array. The microcontroller board provides all hardware necessary to control the system, analyze data and generate alert signals to warn users of adverse conditions. The iEGAS system can communicate with a computer through a USB port or with a mobile device through a plug-in wireless communication module. The package enclosure houses an air inlet, a tiny fan, and particle filters to control airflow across the gas sensor array. The iEGAS system is powered by a rechargeable battery which can be recharged through the USB port.

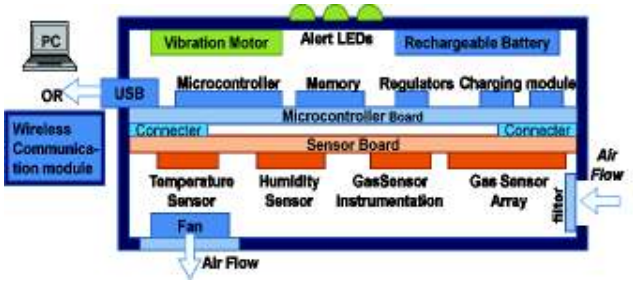

Fig. 1 Illustration of iEGAS gas monitoring device, A roughly thumb-drive sized autonomous sensing system with measurement, analysis, alert and communication capabilities.

The proposed MEIC CMOS instrumentation chip includes two main blocks: signal generators and a multi-mode readout circuit. Fig. 2 shows the functional block diagram of the MEIC and its connection to other iEGAS components. To generate EC stimulus signals, an AC sinusoid generator and DC waveform generator are provided for impedance and amperometry measurements, respectively. The on-chip potentiostat array provides eight-channel variable sensor operating potentials. The multi-mode readout circuit amplifies and records sensor responses and performs analog to digital conversion with very low power and compact size. The MEIC also includes a digital control and communication block and on-chip memory in order to receive commands from and deliver digital data to the iEGAS microcontroller.

IV. DESIGN RESULTS

An IL-based EC sensor was built to verify selected the sensor technolgy. POREX® porous PTFE was used as the permeable membrane for its excellent inertness to electrolyte. Gold electrodes were deposited on the PTFE in an interdigitated manner to maximize the reaction current. High-purity room-temperature ionic liquid $[C_4mpy][NTf_2]$ was used as the electrolyte. The sensor structure and electrode geometry are shown in Fig. 3. EIS measurements were performed for SO_2 as an example pollutant. The impedance response curve in Fig. 4 shows that $\Delta|Z|$ increases with SO_2 concentration. In future work, the sensor structure in Fig. 3

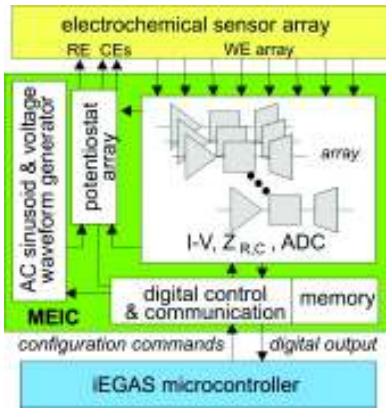

Fig. 2 Block diagram of MEIC instrumentation chip for sensor arrays.

(a) (b)

Fig. 3. (a) EC sensor structure; (b) microfabricated electrode geometry. The electrodes and pads size is 0.75" × 0.5".

will be expanded to an array suitable for a wide range of hazardous gases.

A prototype iEGAS system has been constructed and is shown in Fig. 5. The microcontroller board houses an ultra low power MSP430 (Texas Instruments), memory for data collection, and a battery interface. Humidity and temperature sensors were implemented within the sensor board. The fan and filter were mounted to the system package. This first-generation system is fully operational, able to read sensor data and upload it in real time to a PC for display and storage. The IL gas sensor array and MEIC discussed above will be incorporated into the next generation system.

V. CONCLUSION

The development of a wearable, low-cost, low-power, autonomous, real-time, multi-analyte gas sensor array system, from concept to prototype has been reported. Comparison of gas sensor techniques led to the choice of an electrochemical sensor featuring ILs to provide small size, high sensitivity, good selectivity, rapid response, low power consumption and low cost. A prototype ILs-based EC sensor was built to demonstrate this chosen design concept. To provide low-power, small-size instrumentation for the electrochemical sensors, a CMOS circuit was described that includes multiple EC stimulus techniques, potentiostat biasing, current readout, and A/D conversion. Utilization of signal processing techniques within the miniaturized wearable system was also discussed for optimizing selectivity of the gas sensor array in the presence of a real-world mixed gas environment. An operational first generation prototype integrating the system-level concepts was described and shown. All the components necessary for monitoring hazardous gases were fit into a thumb-drive package with a USB port. Future generations of the system are expected to provide a valuable new tool for the assessment of personal exposure to environmental and occupational hazards.

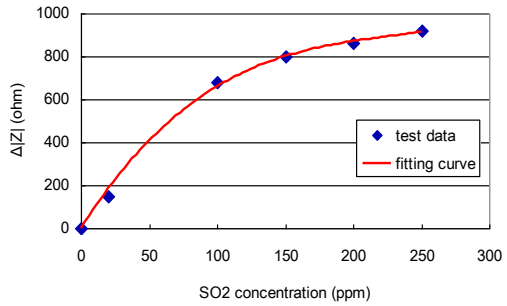

Fig 4. EIS test results plot for different SO_2 concentrations. The sensor operation potential was set at -0.5V and the stimulation frequency was set at 10Hz.

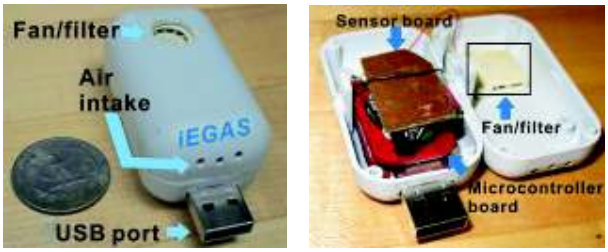

Fig 5. iEGAS prototype system(a) Outer view; (b) Inner view. Humidity and temperature sensors were placed on the sensor board. An MSP430 microcontroller was placed on the MCU board to turn on/off the fan can communicate to remote systems.

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