

Exploitation of spatiotemporal information and geometric optimization of signal/noise performance using arrays of carbon black-polymer composite vapor detectors

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Accepted 22 October 2001

Abstract

We have investigated various aspects of the geometric and spatiotemporal response properties of an array of sorption-based vapor detectors. The detectors of specific interest are composites of insulating organic polymers filled with electrical conductors, wherein the detector film provides a reversible dc electrical resistance change upon the sorption of an analyte vapor. An analytical expression derived for the signal/noise performance as a function of detector volume implies that there is an optimum detector film volume which will produce the highest signal/noise ratio for a given carbon black-polymer composite when exposed to a fixed volume of sampled analyte. This prediction has been verified experimentally by exploring the response behavior of detectors having a variety of different geometric form factors. We also demonstrate that useful information can be obtained from the spatiotemporal response profile of an analyte moving at a controlled flow velocity across an array of chemically identical, but spatially nonequivalent, detectors. Finally, we demonstrate the use of these design principles, incorporated with an analysis of the changes in detector signals in response to variations in analyte flow rate, to obtain useful information on the composition of analytes and analyte mixtures. © 2002 Elsevier Science B.V. All rights reserved.

Keywords: Sensor arrays; Spatiotemporal effects; Conducting polymer vapor sensors

1. Introduction

Arrays of vapor detectors that rely on the partially selective interaction of vapors with various polymers have received significant attention in the recent literature. Detector types of interest include carbon black-insulating polymer composites [1], conducting organic polymers [2–4], polymer-coated quartz crystal microbalances (QCM) [5], polymer-coated surface acoustic wave (SAW) devices [6,7], polymer-coated capacitors [8], and arrays of dye-impregnated polymeric beads or coated optical fibers [9–11]. The responses of such sorption-based detectors depend primarily on the partition coefficient of the gaseous analyte into the polymer [12]. Arrays of detectors, in which each element contains a chemically different polymer, have been demonstrated to allow discrimination between various vapors based on the differences in response patterns produced by the detector array [13].

In most studies to date, the detectors in such an array are placed in nominally spatially equivalent positions relative to the analyte flow path [1,11,14]. In such a configuration, any spatiotemporal differences between detectors are minimized, and the array response pattern is determined by the differing physicochemical responses of the various detectors towards the analyte of interest. The variations in analyte sorption amongst various detectors thus determines the resolving power of the detector array and determines the other performance parameters of such systems. In this work, we have deliberately placed detectors in spatially nonequivalent positions relative to the flow path of the sampled analyte. We demonstrate that the spatiotemporal response properties of such an array can be used advantageously to obtain information on the identity of analyte vapors and also to produce information on the composition of analyte mixtures.

Additionally, in most studies of detector arrays to date, the form factor of the individual detectors is constrained by factors related to the mode of signal transduction. For example, most film-coated QCM devices must have specified dimensions so that a resonant bulk acoustic wave

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can be maintained in the quartz crystal transducer element [15,16]. Similarly, the geometry of SAW devices is constrained by the need to sustain a Rayleigh wave of the appropriate resonant frequency at the surface of the transducer crystal [15]. Each detector in a QCM or SAW array typically has an identical area and form factor; consequently, the array response is based solely on the different polymer/analyte sorption properties of the differing detector films. Although in principle these types of devices could be constructed with a range of form factors, relatively little attention has been focused on varying the form factors of the detector to optimize the signal/noise ratio (S/N) for a particular analyte. Recent work in our laboratories has focused on the use of chemically sensitive vapor detectors comprised of regions of electrical conductors interspersed amongst regions of insulating organic polymers [1]. The swelling of these films upon sorption of an analyte vapor produces a readily measured, dc electrical resistance change. Spray-coating deposition techniques using masked substrates permits the fabrication of such chemiresistor-type vapor detectors in virtually any geometry where the film can bridge two electrically conducting contact leads [17]. This freedom to explore various form factors allows convenient exploration of the geometrical aspects of sorption-based vapor detector design.

We demonstrate herein that different form factors of a given detector film in conjunction with specific types of analyte flow paths can provide very different detection performance for different types of analyte vapors. An analytical expression has been derived to predict the optimum volume of a detector film as a function of the sample volume and the analyte/polymer partition coefficient. Under certain conditions, detectors of very small areas are expected to have the best S/N performance, whereas for other conditions, relatively large detector areas are optimal. These predictions have been verified through measurements of the response properties of conducting polymer composite chemiresistor vapor detectors. We also demonstrate that, based on these principles, the use of an array of detectors that are nominally identical chemically, but which have different form factors relative to the analyte flow path, can provide useful information on the composition and identity of an analyte vapor. Finally, we report S/N data that allow comparisons between the detection limits of several polymer/analyte combinations using two different modes of signal transduction: frequency shifts in SAW devices and dc electrical resistance changes in composites of carbon black and insulating organic polymers.

2. Theoretical considerations

2.1. Dependence of the noise power on the area of a carbon black-polymer composite vapor detector

At open circuit, resistors exhibit voltage fluctuations whose power spectrum is constant as the frequency is varied.

These fluctuations are known as Johnson noise. The root mean squared (rms) noise voltage density of this Johnson noise, V_{JN} , is related to the resistance, R , of a resistive detector as follows:

$$V_{\text{JN}} = (4kTRB)^{1/2} \quad (1)$$

where k is Boltzmann's constant, T the temperature (in K), and B is the bandwidth [18]. This Johnson noise is the fundamental lower limit on the noise of any device of resistance R , and its magnitude is independent of the volume or of other fabrication-dependent properties of the resistor. However, when current flows through most types of resistive materials, a voltage fluctuation is observed with a power spectral density that displays an inverse dependence on frequency. This additional noise, which is typically of the form $1/f^\gamma$, where $\gamma = 1 \pm 0.1$, is designated $1/f$ noise [19,20].

Even for a series of resistors that are fabricated by an identical process, the magnitude of the $1/f$ noise depends on the volume, \mathcal{V} , of the resistor. When the correlation length of the resistive particle network is small compared to the physical length scale of interest, the $1/f$ noise of a resistance-based detector is expected to be proportional to $\mathcal{V}^{-(1/2)}$ [21]. For a given film thickness, this implies that the total noise of a resistive detector scales as $A^{-(1/2)}$, where A is the total area of the detector film between the electrical contact leads. This dependence requires that the magnitude of the $1/f$ noise, in the frequency window of the measurement, is much greater than the magnitude of the Johnson noise, so that the total noise is dominated by the $1/f$ contribution. As a consequence of Ohm's law, the power spectral density, $S_n(V)$, of the $1/f$ resistance noise scales with the square of the bias voltage, V_b , applied to the resistor. The quantity of fundamental interest in characterizing the noise of a resistive detector element is thus

$$S_n = \frac{S_n(V_b)}{V_b^2} \quad (2)$$

where S_n is the relative noise power spectral density and V_b is the biasing voltage [21,22]. In contrast to the Johnson noise, the level of the $1/f$ noise in carbon black-polymer composite resistors varies with many factors, including the structure of the carbon black, its volume fraction in the composite, the type of insulator, the resistivity of the composite, and the method of resistor preparation [21,23].

2.2. Dependence of signal/noise on the area of a carbon black composite chemiresistor

Given the above expectations for the scaling of the noise power of a chemically sensitive resistor with the volume of the detector film, we now consider how the signal produced by sorption of an analyte will depend on the volume of the detector film. Consider introducing a fixed quantity of an analyte into a sample chamber of total volume \mathcal{V}_1 to produce an initial analyte concentration C_v^i in the vapor

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