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Fourier transform mechanical spectroscopy of micro-fabricated electromechanical resonators: A novel, information-rich pulse method for sensor applications

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ABSTRACT

A new sensing method employing a computer-designed broadband voltage pulse sent to an electromechanical resonator is suggested and demonstrated. The pulse spans a user-defined bandwidth around a user-selected center frequency so as to include at least one mechanical resonance. The transient voltage pulse excites mechanical resonance through the piezoelectric effect. The current response signal from the device is recorded and subjected to FFT. Components of the response due to electrical impedance and mechanical resonance are separated and tracked with time upon exposure to solvent vapors, helium, or CO_2. Data are shown for both polymer coated quartz tuning forks and piezoelectric microcantilevers with sub-ppm sensitivity for some analytes.

1. Introduction

Since the introduction of the electronic nose concept by Dodd and Persaud in 1982 several competing sensor platforms based on different physical phenomena have been developed [1,2]. Metal oxide sensors have been used to sense volatile organic carbon in the sub-ppm range as the oxide layer's resistance changes [3], and polymer-based resistive sensors have also been successful [4]. Each of these uses changes in the sensor's electrical resistance caused by analyte adsorption as the basis of sensing. In addition to resistive sensors, the viability of micro-machined mechanical resonators as robust sensors has been recognized for at least 15 years [5,6]. Common resonators include micro-fabricated and nano-fabricated structures made of silicon or quartz [7–9]. Actuation and detection schemes can include optical, magnetic, and purely electrical pathways. Common examples include capacitive detection, piezoresistive electrical detection, and optical detection via laser beam deflection [7,10,11]. The measurement scheme can employ a chirped excitation pulse, a phase locked loop, a broadband pulse, or even intermittent excitation at a single frequency near resonance [12–15]. A high-frequency, multi-frequency method has been developed by Roukes et al. for NEMS resonators with frequencies in the 10 s or 100 s of MHz using a magnetic field to transduce the actuation signal [14]. Kasemo and Rodahl used intermittent excitation near resonance and analysis of the resulting "ring down" to give both the resonance frequency and the quality factor, but no purely electrical information was extracted thusly [15].

The resonators are typically coated with a chemically sensitive material that is either uniquely specific or moderately specific. Common coatings include antibodies, mesoporous materials, metal oxide films, and various polymers with each class of coating aimed at a specific sensing application [16–19]. Systems employing moderately specific coatings like mesoporous, metal oxide, or polymer films compare data from an array of sensors to enhance specificity with a sort of fingerprint technique where the combination of moderately specific responses is unique to an analyte.

Here we present a fundamentally new detection scheme based on excitation of an electromechanical resonator with a single transient electrical pulse and immediate, pre-programmed computer-based analysis of the transient response. The response current is recorded and Fourier transformed. Then it is decomposed into mechanical resonance and standard electrical impedance com-

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ponents, and each is fit to the expected peak shape based on simple physical models. The mechanical resonance, f_0; the mechanical quality factor, Q; the amplitude of the mechanical resonance, a; the capacitance, C; and the transmitted electrical phase shift, Φ, are then recorded over time as the oscillators are exposed to analyte vapors. Using 32.768 kHz resonators, the system measures at roughly 1 pulse per second, so that an array of 8 multiplexed sensors updates roughly every 10 s. Since this timescale is comparable to that of the diffusion of trace analytes in common fluids (water and air) under ambient conditions, the measurement may be considered real-time. Unlike many of its competitors that require bulky optics setups or external magnetic fields, this pulse-response method uses a purely electrical measurement scheme. In addition, this method eliminates the need for a chirped excitation signal in the measurement circuit, allowing much faster measurement times. The user-defined, computer-generated broadband voltage pulse ensures consistent excitation and allows easy decomposition of the response signal into electrical and mechanical components. So the capacitance of a resonator and the phase of the circuit can be tracked through successive excitations at the same time that measurements of resonance frequency, quality factor, and electromechanical coupling are made. The end result is more independent information gathered in a much shorter time than is currently possible.

This information can be used to distinguish analytes based on several different physical phenomena simultaneously. For example, the resonance frequency and quality factor of a rectangular beam clamped at one end with a homogenous adsorbate are described by Eqs. (1) and (2) [20]:

$$\frac{f^{(n)}}{f_o^{(n)}} = \sqrt{\frac{D/D_o}{1+\rho t}} \tag{1}$$

$$\frac{Q^{(n)}}{Q_o^{(n)}} = (1+\rho t)\frac{\mathrm{Im}\ \Gamma(f_o^{(n)})}{\mathrm{Im}\,\Gamma(f^{(n)})} \tag{2}$$

Eq. (1) gives the relative shift in resonance frequency in terms of the mass change (ρt) and the change in flexural rigidity, D, which depends on the thickness (t) and the Young's moduli of the adsorbate (E_a) and the cantilever (E_c). Eq. (2) gives the relative shift in the quality factor in terms of the mass change (ρt) and the hydrodynamic function ($\Gamma(f)$) for a rectangular beam at a given frequency. Interestingly, Grüter and colleagues showed that it is not possible to use the frequency shifts alone to determine change in mass or rigidity or both. However, knowledge of the resonance frequency and quality factor before and after exposure is sufficient to determine both the change in mass and the change in rigidity [20]. Fortunately, this method gives measurements of both resonance frequency and quality factor with each pulse, enabling the simultaneous, accurate determination of both change in mass and change in flexural rigidity when the surrounding fluid properties are known.

The resonators are coated with various polymer films to allow some degree of chemical specificity to the resonator–analyte interaction. Polymers are chosen to match previously published results and to include at least one water-soluble and one water-insoluble. The magnitude of the mechanical response represents electromechanical coupling and it is a complicated function of polymer film stiffness, surface charge effects, and surrounding fluid properties. Capacitance depends on the dielectric constant of the polymer film, which changes as polar or charged functional groups on the analytes interact with the polymer matrix. The phase of the electrical current response has several dependences including the measurement electronics, and it is demonstrated to change in response to analytes. These five independent variables describe the interaction of a single analyte with a single polymer film, making this information-rich technique more capable of distinguishing analytes.

2. Materials and methods

2.1. Overview

The experimental setup is shown in Fig. 1. A voltage pulse is specified using the arbitrary waveform VI in Labview 8.5 and output from National Instruments DAQ USB-M6259 BNC. The voltage pulse is carried through standard BNC cables to a home-built multiplexing circuit that allows us to address each of the resonators individually. The response current from the resonator passes through the multiplexer to an SR 570 current pre-amplifier in broadband mode where it is converted from a current to a voltage and amplified with a gain of 10^5 V/A. Finally, this response voltage signal is cabled to an analog in port on the DAQ hardware.

2.2. Resonators

Individual resonators were soldered into standard IC chip sockets that were plugged into a home-built multiplexing circuit. The resonance frequency was measured independently using a NanoSURF EASYPLL in controlled amplitude mode for comparison.

2.2.1. Polymer coated quartz tuning forks

Quartz tuning forks, with nominal resonance frequencies of 32,768 Hz ±20 ppm were purchased from Frequency Management International (FMI). The metal capsules were removed so that both arms of the tuning fork were exposed to the surrounding atmosphere. Fig. 1 includes a photograph of a quartz tuning fork, note that it comes with thin metal electrodes deposited in a pattern on the surface and connected to two wire leads by solder at the base. The tuning forks were then dip coated in a 2.9% (w/w) aqueous solution of poly(ethyleneimine) (PEI), wicked between the prongs with a Kimwipe, and allowed to dry under ambient conditions as shown in Fig. 2. Other polymer coatings were introduced by the same method with poly(aniline) (PAn) in DMSO and poly(vinyl alcohol) (PVA) in H_2O.

2.2.2. Piezoelectric microcantilevers

Piezoelectrically AlN-driven silicon-based microcantilevers were used. The silicon cantilever base had a thickness of 20 μm

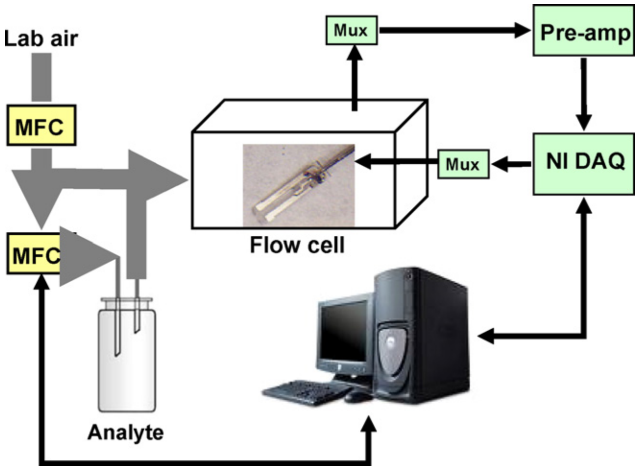

Fig. 1. Overall experimental schematic: An electronic signal is sent from the control electronics to a resonator in the analyte chamber. A PC controls a pair of mass flow controllers that introduce a known quantity of analyte. The output signal from the resonators is recorded and processed.