Electromechanical film as a photoacoustic transducer

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Abstract: An electromechanical film, EMFi, is utilized as a transducer in a photoacoustic (PA) gas sensor. The film is a sensitive acoustic transducer, it is easily formable, and it exhibits a wide frequency response regardless of its large surface area. As a demonstration of its capabilities, the EMFi-based PA detector is used to measure NO_2 with pulsed excitation at 436 and 473 nm. The minimum detectable absorption coefficient is extrapolated to be $5 \cdot 10^{-7}$ cm^{-1}. Improvements for EMFi-based PA detector are discussed.

OCIS codes: (040.0040) Detectors; (300.6360) Spectroscopy, laser.

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1. Introduction

Trace gas detection is important in medical, industrial, and environmental research and monitoring. Emission standards for industry and energy production plants generate necessity for sensitive gas detectors. In situ gas detection provides valuable information for better understanding and optimizing the industrial processes. Trace gas detection is also a noninvasive diagnostic method for medical science. Photoacoustic spectroscopy (PAS) is generally considered as one of the most sensitive gas detection methods. A resent development of inexpensive but powerful laser light sources as well as a relatively simple implementation of the photoacoustic (PA) instrumentation makes PAS-based gas detector attractive for different applications. PA gas detector can also be used for in situ, real-time gas analysis during the industrial process [1–3]. Detection principle of the PA detectors is based on an excitation of the sample by the light and a detection of the thermal relaxation-induced pressure difference. The detection of the pressure difference, i.e. an acoustic wave, is based on an acoustic transducer. Conventionally, a microphone is used as the transducer. The microphone acts as a point detector for the pressure signal. However, the laser induced acoustic wave possess a cylindrically shaped wavefront, therefore, the planar microphone is not shaped optimally for the detection of the acoustical wave. Exploiting the acoustical resonances of the cylindrical gas cell require high signal frequencies up to tens of kHz to be detected. Some suggestions for the detection of cylindrical waveforms are published in literature. A tubular array of up to 80 microphones is used with a cylindrical PA cell [4,5]. In another application, an electret foil was wrapped around a metal tube, having closely spaced holes [6]. The Q value of the machined cell was around 30 at the resonance.

In this work, electromechanical film (EMFi), also referred to as EMFIT film [7,8], is used as a PA transducer. EMFi is thin and flexible cellular polypropylene film, having permanent internal charge. When external force is introduced onto the EMFi, an induced mirror charge is collected from metalized surfaces of the film. EMFi is durable, inexpensive, formable, and has a very wide frequency response of up to hundreds of kHz. Stacking films together increases sensitivity of the transducer [9]. Formability of the film enables utilization of a large area cylindrical acoustic transducer. An acoustic noise of a turbulent gas flow is mostly occurring below 2 kHz [10]. The use of flowing samples is possible with EMFi, due to the wide frequency response of the film. As a demonstration of its capabilities, a detection limit of 27 ppb of NO_2 in nitrogen is extrapolated from flowing gas measurements.

2. Electromechanical film in photoacoustics

The EMFi is typically 70 µm thick polypropylene film with voided internal structure. The film is corona charged during the manufacturing process. The permanent charges are located on the surfaces of the air voids. Both sides of the film are metalized to form electrical contacts. As an external force is introduced to the film, the voids are compressed and the opposite charges are brought closer together. Thus, a mirror charge is collected from the metalized surfaces. The electromechanical effect is related to the voided structure of the EMFi, rather than the piezoelectricity of the material itself. The sensitivity of the EMFi is often expressed with the piezoelectric coefficient d_{33} in pCN^{-1} units [7]. The porous structure of the film enables sensitivities up to 790 pCN^{-1} [11]. The sensitivity of the EMFi, used in this study, is estimated to be about 140–150 pCN^{-1}. The sensitivities of conventional piezoelectric films like PVDF, PZT-polymer composite and ceramic PZT are in the range of 20-600 pCN^{-1}, whereas the coupling factors of the piezoelectric films are higher than for EMFi [12]. An acoustic impedance of EMFi is two orders of magnitude smaller than that of traditional piezoelectric films and thus it is better matched with the ambient air. A figure of merit, describing the efficiency of a transducer film in air-coupled applications, is 165 for EMFi, 42 for composite PZT and less than 1 for PVDF and ceramics PZT [12]. The reported sensitivity of EMFi is 2.2 $mVPa^{-1}$ [9], which is comparable to 1 $mVPa^{-1}$ of commercial (B&K type 4138) 140 kHz frequency bandwidth condenser microphone. The sensitivities of commercial Knowles electret microphones are in order of 10 $mVPa^{-1}$, whereas the bandwidth of this type of microphones is limited to 10-20 kHz.

Previously EMFi was used for ultrasonic applications in air [12,13], for an artificial bat head [14], for keyboards, loudspeakers, medical monitoring, and a number of other applications [7]. The diversity of the applications can be explained by the properties of the film. The film is flexible, durable, sensitive, and has wide frequency response up to hundreds of kilohertz. The sensitivity can be further enhanced by stacking the films together. The frequency response of the EMFi is flat and wide. A single-layer film has resonance frequency at about 300 kHz, whereas the resonance frequency of the double-layer stack is reduced to about 70-90 kHz [15]. Increasing the size of the transducer increases the total detected acoustical energy. The size of traditional microphones is fixed, whereas the size of the EMFi can be altered without a reduction of the frequency bandwidth. The large size of the film can be exploited in PA applications to maximize the transducer area inside the PA cell.

The laser beam induces a cylindrically shaped acoustic wave. Thus, cylindrical cells are often used in PA applications, especially when acoustical resonances of the cell are exploited for the signal amplification. In this work, a simple 100 mm long iron tube with 18.5 mm radius is used as PA cell. Windows are placed at the ends of the tube. A two-layer EMFi is glued inside the tube covering half of the cylinder, as is shown in Fig. 1. To be able to measure azimuthal modes of the cell, the second half of the inner surface is left bare. Since the best signal to noise ratio (SNR) in our case is achieved with radial resonances, the azimuthal modes are only used for aligning the laser beam. Minimizing the azimuthal modes ensures centrosymmetric beam path inside the PA cell. The sensitivity of the detector could be nearly doubled just by coating the whole inner surface of the tube with the film. The EMFi transducer is attached to the cell with epoxy glue, whereas the films are attached to each other and to electrical contacts with a conductive epoxy. A self-made amplifier is used to amplify the output current signal of the film. The amplified signal is averaged and recorded with a digital oscilloscope (WaveRunner6100A, LeCroy) for further data processing.

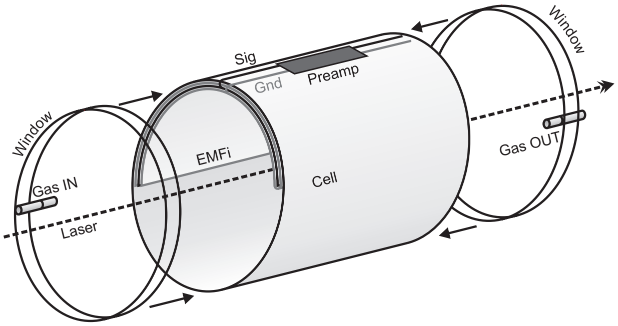

Fig. 1. Photoacoustic cell with EMFi transducer. An upper half of the inner surface of the iron tube is coated with a double layer EMFi. Ground (Gnd) and signal (Sig) contacts to preamplifier (Preamp) are located on the top of the cell.

The performance of the PA instrumentation was demonstrated with 5–45 ppm of NO_2 in nitrogen as a sample gas at atmospheric pressure. The sample gas of 186 ppm of NO_2 in nitrogen was diluted with pure N_2 by a mass flow controller (5850S, Brooks Instrument). A pulsed OPO laser (NT342/1/UVE, Ekspla) operating at 10 Hz repetition rate, having the tuning range of 210–2300 nm, was used as a light source. Excitation wavelengths of 436 nm

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and 473 nm were used with average pulse energies of 5.8 mJ and 4.6 mJ, respectively. The pulse duration of the laser was 5 ns. The laser beam diameter was about 1 mm. The short laser pulse excites a broadband acoustical pulse [16], thus the characterization of the cell resonances was performed with a pulsed excitation.

3. Results and discussion

An averaged photoacoustic signal from 5 ppm of NO_2 in N_2 both in time and in frequency domains are shown in Fig. 2. The detectable acoustic wave, shown in the inset of Fig. 2, resonates in the PA cell for about 10 ms. The Q-switched laser produced a high amplitude electrical noise at the beginning of the signal, thus a smooth but stiff window function was used to clean the beginning of PA signal during the signal processing. Also PA signal after 10 ms was reduced with exponentially decaying window function. PA signal was then normalized with the corresponding average pulse energy and transformed into the frequency domain with Fourier transform (FT). The dumped acoustic resonance vanishes from the recorded signal after about 10 ms. Therefore, a zero padding property of FT was applied to continue the time domain signal with zeros to 200 ms. The frequency range of the detector was mostly limited by the self-made preamplifier. Therefore, the highest SNR of the responses is located at around 60 kHz. The Q-value for the 6th radial resonance at 59.2 kHz is over 1180 when the Q-value is defined as the frequency of the mode divided by the full width at half-maximum, FWHM of power spectrum [17]. First ten radial modes are labeled in the Fig. 2 and can clearly be distinguished below 100 kHz. Other peaks in the frequency spectrum in between the radial modes are azimuthal, longitudinal and combinations of the resonance modes. Existence of these extra modes is originated from the unideal shape and direction of the laser beam.

The recording time of an individual signal was 20 ms. However, due to the static electrical noise, the time frame of the signal was windowed to about 10 ms. 100 signals were averaged in each measurement, resulting in a nominal measurement time of 1 s or a real measurement time of 10 s, if 10 Hz repetition rate of the laser is taken into account.

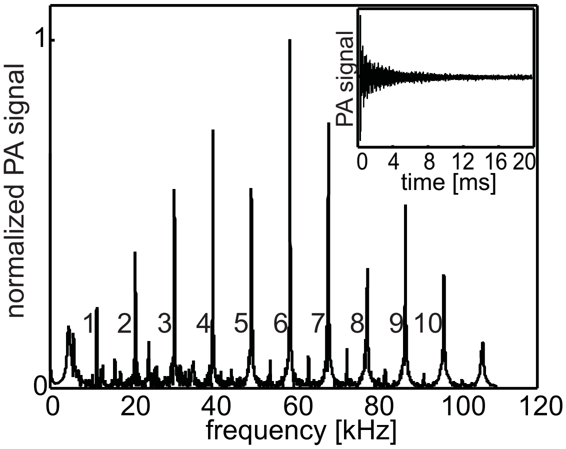

Fig. 2. Averaged PA signal of 5 ppm NO_2, excited at 473 nm. Main figure: frequency domain, first ten radial eigenmodes labeled. Inset: PA signal in time domain.

PA signal is linearly dependent on the sample gas concentration. The linearity of our detector was measured at the concentrations from 5 ppm to 45 ppm. Nitrogen dioxide tend to stick on the surfaces, thus a high total flow of 5 $Lmin^{-1}$ was used. Since the acoustical noise from airflow is generated well below 10 kHz, the PA measurements at around 60 kHz were not affected by the acoustic noise. The measured PA signal as a function of concentration is shown in Fig. 3. The signal levels are determined from the peak of the 6th radial mode. The measured signals show linear dependence of the PA signal on the sample gas concentration within the measured range. We were unable to reliably generate sample gas with concentrations less than 5 ppm, thus the sensitivity limit of the instrumentation had to be extrapolated from the presented data. The 42 ppb sensitivity limit of the instrumentation is reached at the crossing point of the linear fit and the rms noise level of the 6th radial mode. For the noise level measurements, PA cell was washed with pure nitrogen for several minutes. The noise level was determined by calculating rms values of 20 PA measurements, each having 100 averaged signals. All the measurement conditions were kept similar to the measurements of the NO_2 samples.

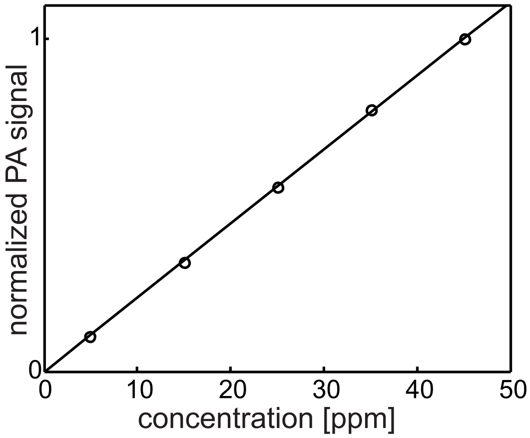

Fig. 3. Concentration measurements of NO_2 at 473 nm excitation. Circles: measured amplitudes of the 6th radial mode at 59.2 kHz, line: linear fit. Detection limit corresponding to $S/N_{rms} = 1$ is 42 ppb.

The absorption cross-sections of NO_2 [18] and the average pulse energies used are listed in Table 1. The measured SNR of the 6th radial mode from 5 ppm NO_2 at 436 nm is over 180, which correspond to 27 ppb detection limit of the instrumentation. Further lowering the wavelength is a trade-off between increasing the absorption cross-section and decreasing the pulse energies of our laser without overall SNR improvement. The sensitivity of the presented PA detector can be expressed as a minimum detectable absorption coefficient of $\alpha_{min} = 5 \cdot 10^{-7}$ cm^{-1}.

Table 1. Properties of NO_2 and corresponding laser pulse energies

Laser wavelength [nm]	Absorption cross-section [cm^2]^a	Average pulse energy [mJ]
436	$7 \cdot 10^{-19}$	5.8
473	$4 \cdot 10^{-19}$	4.6

[a] Literature values [18]

In this study, the performance of EMFi is demonstrated with a high-frequency PA cell. The presented instrumentation was designed to show the capabilities of the new transducer, rather than to optimize the gas detector. The sensitivity of EMFi-based detector can be improved in several ways. Highest reported sensitivity of EMFi is 790 pCN^{-1} [11], which is almost 6 times larger than the one used in this study. The whole inner surface of the resonator tube should be covered with the film, and also prolonging the resonator will increase the surface area of the EMFi. Stacking the films together will increase sensitivity and lower the resonance frequency of the transducer, if lower frequencies are preferred [9,15]. It would be advantageous to use the resonance frequency of the EMFi to maximize the signal. Proper focusing of the laser beam will produce the initial acoustic pulse with a peak frequency at the desired band [16]. The diameter of the PA cell can be fixed to produce 1st radial resonance at the same frequency. The pressure amplitude at the walls of the cylindrical PA cell is about 40% of the pressure amplitude in the middle of the cell for the 1st radial resonance mode [17].

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The pressure amplitude at the walls is only 30% for the 2nd radial mode, 25% for the 3rd and is further reduced for higher resonances. Thus, the first radial mode is often preferred.

Some properties of EMFi are to be studied. The frequency response and SNR improvement of EMFi-based PA detector, as the film size and a number of layers in a stack are increased. Different cell geometry could be introduced to utilize multipass or cavity enhancement of excitation. Because EMFi is a whole new kind of acoustic transducer in gas phase PAS, fresh ideas for PA cell geometry can be implemented.

4. Conclusions

We have utilized EMFi transducer in PA detector for the first time. EMFi is flexible, durable, inexpensive, and regardless of its large size, combines relatively good sensitivity and a wide frequency response of hundreds of kilohertz. Compared to traditional microphones, formability of the film enables optimal detection of cylindrical acoustic waves inside the acoustic cell. The minimum detectable absorption coefficient of the presented gas detector is $\alpha_{\min} = 5 \cdot 10^{-7}$ cm^{-1} for NO_2 in nitrogen at the atmospheric pressure and 436 nm excitation wavelength, which corresponds to the detection limit of 27 ppb. As the film is easily formable and stackable, geometry of PA detector can be completely rethought to take full advantage of the new transducer material.

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