

# Distinguishing Chemicals Using CMUT Chemical Sensor Array and Artificial Neural Networks

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**Abstract**— Capacitive micromachined ultrasonic transducers (CMUTs) can function as extremely sensitive mass-loading chemical sensors. The resonant frequency of the CMUT changes as mass is added due to chemicals absorbing into a chemical-sensitive layer on the top of the plate. However, these sensors suffer from the problem that they are not selective to a single chemical. As a solution, we present a system of four CMUT chemical sensors with different functionalization layers. Neural networks are used to do pattern recognition on the sensor outputs in order to distinguish different chemicals. The system is capable of distinguishing water, ethanol, acetone, ethyl acetate, methane and carbon dioxide in air at concentrations less than 1% with 98% accuracy. Once the chemical is identified, the concentration can be determined using polynomial regression with an RMS percentage error ranging from 1.1% to 13%, depending on the analyte.

**Keywords**—CMUT, Chemical Sensor, Neural Network, Machine Learning

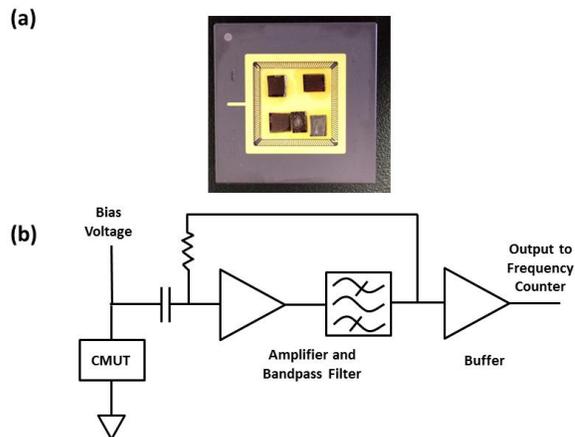
## I. INTRODUCTION

Capacitive micromachined ultrasonic transducers (CMUTs) can function as extremely sensitive chemical sensors [1]. The sensors work using a mass-loading mechanism. Chemicals are reversibly absorbed into a chemical-sensitive coating deposited on the top of the CMUT plate. The increased mass due to the bonded chemicals shifts the resonant frequency to undergo a fractional frequency shift given by

$$\Delta f / f = -\Delta m / 2m \quad (1)$$

where  $m$  is the effective mass of the CMUT plate. Previous work has demonstrated the ability of the CMUT to detect a variety of chemicals. For example, detection of dimethyl methylphosphonate, a sarin gas simulant has been demonstrated with a limit of detection of 50.5 parts per trillion [2].

As microelectromechanical systems (MEMS) devices, CMUTs have many advantages, such as small size and low cost at high volumes. However, CMUT chemical sensors lack selectivity, as is typical of MEMS chemical sensors relying on reversible absorption of the analyte into a functionalization layer. The functionalization layers display some sensitivity to



**Fig. 1:** (a) Five chips containing CMUTs on a chip carrier. Four are used in these experiment: three with functionalization layers and one with none. (b) Schematic of the oscillator circuit. The resistor and CMUT form a voltage divider which has zero phase and a local maximum in gain at the CMUT's parallel resonant frequency. With the amplifier and bandpass filter, the loop has gain  $> 1$  and zero phase shift at the CMUT parallel resonant frequency, allowing stable oscillation.

many different chemicals. This problem can be overcome by using an array of chemical sensors with different sensitivities. Chemicals or characteristic scents can then be recognized using statistical and machine learning techniques [3]. In this paper, we demonstrate the application of this approach to CMUT chemical sensors. We made a CMUT chemical sensor system consisting of four sensors operated simultaneously. Using an artificial neural network, the system can reliably distinguish between different chemicals at low concentrations ( $< 1\%$ ) in nitrogen. Once the chemical is identified, its concentration can be determined using second order polynomial regression.

## II. THE CMUT CHEMICAL SENSOR SYSTEM

The CMUTs used in this work were fabricated using a local oxidation and wafer bonding process [4]. They have a radius of  $15 \mu\text{m}$ , a plate thickness of  $1 \mu\text{m}$  and a gap height of  $255 \text{ nm}$ . Their resonant frequency is  $14 \text{ MHz}$ . There are 670 CMUT cells per device. The large number of cells is advantageous since it results in a better signal-to-noise ratio than would be possible with a single-cell device.

Chemical	Concentrations Tested (ppm)	Number of Measurements
Methane	199-794	13
Carbon Dioxide	2953-4921	33
Water	20-203	11
Ethanol	38-786	18
Acetone	185-3245	14
Ethyl Acetate	161 - 1608	28

**Table 1:** Summary of the chemicals used in the detection experiments, the concentrations they were used at, and the number of measurements done with each type.

The resonant frequency of each device is measured continuously using an oscillator circuit, as described in [2]. The oscillator circuit, shown in Fig. 1 (b), is designed to oscillate at the parallel resonant frequency of the device, where the impedance magnitude is a local maximum and the phase is zero. A buffer connects each oscillator to a frequency counter (SR620, Stanford Research Systems, Sunnyvale, CA), which measures the frequency.

For these experiments, five chips were attached to a chip carrier as shown in Fig. 1 (a). Each chip contains many devices, but only one from each chip was used. Four of the five chips were used in these experiments.

One device was left with no functionalization layer. Its surface is silicon with a thin layer of native oxide. The remaining three devices were functionalized with propyl gallate, polycaprolactone and poly(ethylene oxide). These chemicals were dissolved in tetrahydrofuran, toluene and isopropanol, respectively, to a concentration of 3 mg/ml. 2  $\mu$ L of the solution was deposited on each device using a micropipette.

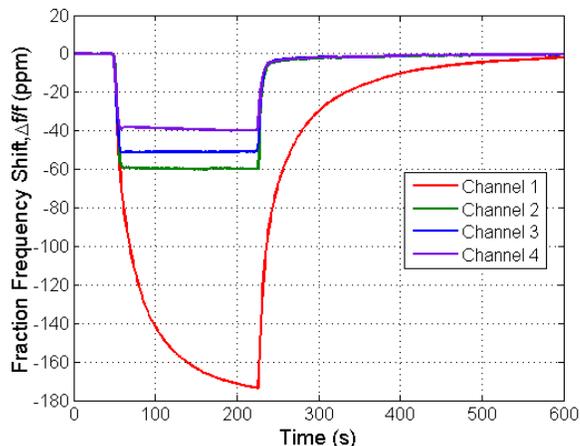
### III. CHEMICAL RESPONSE MEASUREMENTS

The devices were enclosed in a small test chamber with a volume of 10 ml. The test gases were flowed continuously through the test chamber, and the flow rates were controlled using mass flow controllers. The vapors (ethanol, acetone, ethyl acetate, water) were produced using bubblers. Carbon dioxide and a 2% mixture of methane in nitrogen were supplied by gas cylinders. The test chemicals and vapor-saturated nitrogen were flowed at 1-10 ml/min and mixed into 200-500 ml/min flow of nitrogen to reduce the concentration. The chemicals tested and the concentrations in parts per million (ppm) they were tested at are summarized in Table 1.

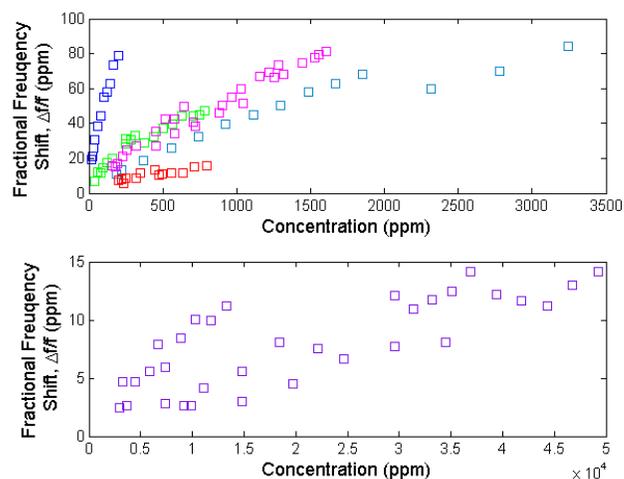
In each chemical detection experiments, nitrogen alone was flowed over the sensor for 45 seconds. Then, the chemical to be detected was flowed over the sensor for 180 seconds, with the diluting nitrogen flow adjusted to keep the total flow rate constant. Finally, nitrogen was flowed over the device again until the sensor returned to equilibrium. Fig. 2 shows an example of the sensor response. Fig. 3 shows the final frequency shift (after three minutes of chemical exposure) of the propyl gallate functionalized sensor versus concentration.

### IV. CHEMICAL IDENTIFICATION WITH MACHINE LEARNING

Two different approaches to chemical classification were tested: linear regression and artificial neural networks. Linear



**Fig. 2:** Response of the system to 2318 ppm of acetone vapor in nitrogen. From 0 to 45 seconds, nitrogen only is flowed over the device. From 45 to 225 seconds, nitrogen with acetone vapor is flowed over the device. After 225 seconds, only nitrogen is flowed over the device. Channel 1 is functionalized with propyl gallate, channel 2 with polycaprolactone and channel 3 with poly(ethylene oxide). Channel 4 is not functionalized.

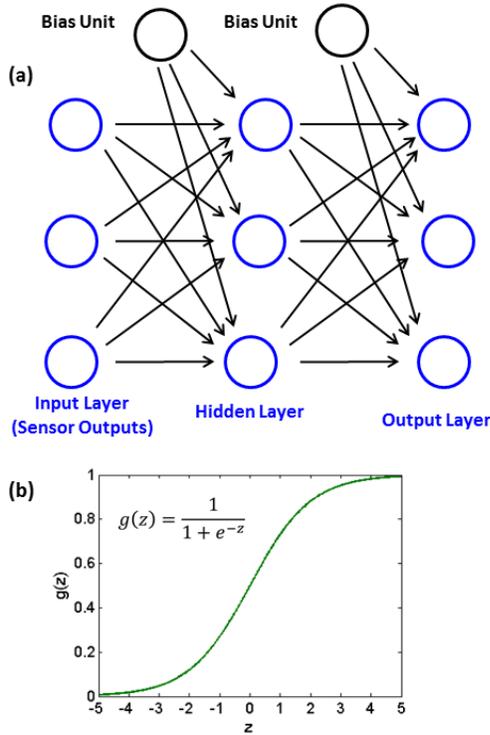


**Fig. 3:** Fractional frequency shift of the propyl gallate functionalized sensor in parts per million (ppm) in response to three minutes of exposure to each analyte

regression is a simple method and provides a benchmark against which the neural network approach can be compared. All of the data processing was done using MATLAB. For both approaches, the pipeline from sensor output to identification and concentration determination is as follows:

1. Preprocessing: remove drift and scale the frequency data so that all points are  $\leq 1$
2. Extract the features to use for classification model
3. Use linear regression model or a neural network to identify the chemical
4. Determine the concentration using second order polynomial regression.

$$y_i = \theta_{i0} + \theta_{i1}x_1 + \theta_{i2}x_2 + \dots \quad (2)$$



**Fig. 4:** (a) Schematic of a simple artificial neural network. The circles represent nodes and the arrows represent connections between nodes. The nodes (other than the input layer) have a sigmoid transfer function as shown in (b). The connections between nodes have different weights. Each node receives a weighted sum of the outputs of the previous layer. The bias units produce a constant output.

CMUT chemical sensors exhibit slow frequency drift caused by things such as changes in temperature or charging of the insulator layer in the bottom of the vacuum cavity below the plate. We correct for this by performing a linear fit to frequency versus time before the test chemical was introduced and long enough after it was turned off that the sensor has returned to equilibrium.

The fractional frequency shifts are normalized so that all points are  $\leq 1$ . This is done because neural networks function best with inputs roughly between -1 and 1.

Next, the features of the sensor responses to be used for identification are extracted. Two different sets of features are used here. The first set consists of the final normalized fractional frequency shift, measured just before the test chemical is turned off. The second set consists of samples taken of the normalized fractional frequency shift every 15 seconds until the test chemical is turned off. The advantage of the second set of features is that it allows the learning algorithm to use the response time of the sensor to aid in identification.

After that, the features are fed into the learning algorithm. For linear regression, the concentration of each chemical is modeled as a linear function of the sensor output features with equations of the form

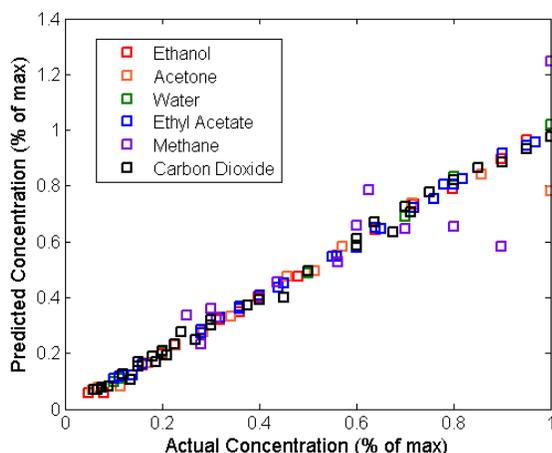
Method	Features Used	Average Training Data Error	Average Cross-Validation Error
Linear Regression	Final frequency shifts	75%	74%
Linear Regression	Final frequency shifts and their squares	87%	86%
Linear Regression	Sample every 15 s	99%	91%
Neural Network (8 node hidden layer)	Final frequency shift	94%	89%
Neural Network (12 node hidden layer)	Sample every 15 s	99%	98%

**Table 2:** Summary of the performance of the chemical identification algorithms.

where  $y_i$  is the concentration of chemical  $i$ ,  $x_j$  are the sensor output features and  $\theta_{ij}$  are the fitting parameters. To use the model for identification, the sensor data is fed into the model and the chemical is identified as the one with the highest concentration. For this model, the vapor concentrations for each chemical are normalized by the maximum concentration of that chemical to prevent a bias towards chemicals tested in a higher concentration range. Linear regression can be adapted to nonlinear situations by including higher order terms as features. In this work, we demonstrate both a linear model and a second order polynomial model where the squares of the sensor outputs are included.

Artificial neural networks are more complex models which have the advantage that they can handle complex, nonlinear relationships between input and output without the model creator having to specify them. Fig. 4 shows a schematic of a simple neural network. It consists of layers of nodes with weighted connections between them. The nodes other than those in the input layer have a sigmoid transfer function, as shown in Fig. 4. Each sensor output is assigned to one of the nodes in the input layer. These values are passed with various weights to the hidden layer nodes, and its outputs are passed through weighed connections to the output layer. The bias units each produce a constant output. The output layer thus produces a value between zero and one. In our model, each node of the output layer corresponds to one of the chemicals that may be present. Using the backpropagation algorithm, the model is trained so that the output node corresponding to a given chemical outputs one if that chemical is the chemical being measured and zero if it is not. In operation, the chemical is identified by choosing the node with the highest score. In the results shown here, the neural network has one hidden layer. However, a neural network may have any number of hidden layers. It should also be noted that neural networks are a large class of algorithms, and the type implemented here is just one version.

To help prevent overfitting, we use regularization, which imposes a penalty on extreme values of the connection weights. To ensure that the models are not overfitted, we cross-validate using a leave-one-out approach. In leave-one-out cross



**Fig. 5:** Concentration predicted by the linear regression model versus the actual concentration of the chemical. Chemical concentrations are plotted as a percentage of the maximum concentration of that chemical tested.

Analyte	RMS Error (%)
Ethanol	1.1%
Acetone	6.1%
Water	1.5%
Ethyl Acetate	1.1 %
Methane	13%

**Table 3:** RMS percentage error in concentration predicted by the linear regression model for each of the chemicals tested.

validation, the model is trained using all but one measurement and then tested on the remaining measurement. The process is repeated, leaving each measurement out in turn.

Table 2 summarizes the performance of each of the chemical identification approaches. The performance of linear regression improves when squares of the sensor responses are included, allowing it to better match the nonlinear dependence of frequency shift on concentration that can be seen in Fig. 3. Linear regression using the frequency sampled every 15 seconds performs slightly better, but the discrepancy between the training data error and the cross-validation error suggests that this model is suffering from overfitting. The neural network using only the final frequency shifts outperforms both of the linear regression models using final frequency shifts. The neural network using frequency samples every 15 seconds gives excellent performance, successfully identifying the chemical 98% of the time.

## V. MEASURING CONCENTRATIONS

Once the identity of a chemical is determined, we find its concentration using second order polynomial regression. Here, we use the final fractional frequency shifts as features. The model was tested using a leave-one-out approach. Fig. 5 plots the concentration calculated by the model vs. the actual concentration for each test. The RMS percent error in the concentration found by the model is shown in Table 3. The method performs well, with accuracies ranging from 1.1% for ethanol and ethyl acetate to 13% for methane.

## VI. CONCLUSION

We demonstrated an array of four non-specific CMUT chemical sensors capable of distinguishing between six chemicals with 98% accuracy. We showed that neural networks used in combination with a CMUT chemical sensor array can be effective in identifying chemicals and can outperform simpler linear regression models. Additionally, we showed that using the response of the CMUT chemical sensor

over time rather than just the final frequency shift can improve classification performance.

Neural networks are one of many machine learning algorithms. Other approaches such as support vector machines would likely also be effective. Machine learning has been used in chemical sensor applications such as breath analysis [5] and tea quality prediction [6]. Machine learning would likely help CMUT chemical sensors to perform well in these types of situations, where a complex characteristic chemical signature must be detected rather than a single chemical. These techniques may also allow CMUT chemical sensors to be effective at sensing multiple components in a chemical mixture, or sensing a single chemical in the presence of interfering chemicals like water vapor. This will be explored in future work.

## ACKNOWLEDGMENTS

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