MULTIVARIATE ANALYSIS OF SH-SAW BASED E-TONGUE SENSORS

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Abstract:
A shear horizontal surface acoustic wave (SH-SAW) resonator sensor operating in the 434 MHz industrial, scientific and medical (ISM) band has been used for identification of basic taste samples. The resonator's capability to differentiate not only between different tastes through their physical properties but also between samples of with different taste intensities has been demonstrated. Acetic acid, sucrose and sodium chloride solutions with 0.5%, 1%, 3% and 5% concentrations were chosen for evaluation and a multivariate analysis was performed on the sensor responses. Linear regression models were fitted to the measured data; the derived linear models were statistically significant offering a simplified alternative to the complex analytical models of the SH-SAW sensors.

Keywords: shear horizontal surface acoustic waves, electronic tongue

NTRODUCTION

There is no unanimity in the scientific community regarding the definition of the electronic tongue (e-tongue) but it usually refers to "an analytical instrument comprising an array of non-specific, poorly selective, chemical sensors with partial specificity (cross-sensitivity) to different compounds in a solution, and an appropriate chemometric tool for the data processing" [1]. Electronic tongue systems are usually based on electrochemical techniques and are composed of several kinds of ion selective electrodes/membranes that convert the physical properties of taste substances into electronic signals. The sensors have different output patterns for chemical substances with different taste qualities [2]. The most common types of taste sensors used are based on electrochemical techniques such as potentiometry and voltammetry [3-4].

The work described in this paper is based on a shear-horizontal surface acoustic wave (SH-SAW) sensor with no bio-chemical layers, thus relying on a physical, rather than electrochemical principle of operation. The resulting device is not just smaller in size but also is more robust and durable than electrochemical sensors.

E-TONGUE SH-SAW SENSOR

Shear horizontal surface acoustic wave (SH-SAW) delay line devices have been successfully used as liquid sensors at Warwick University and their performance as electronic tongue analytical systems has been previously reported [5-6]. The SH-SAW electronic tongue systems classified correctly the different basic tastes *without* a selective biological or chemical coating.

The sensors described here belong to the second generation of coating-free SH-SAW devices that operate at higher frequencies in the 434 MHz ISM band. The sensor employed in this work has a two-port resonator configuration with an electrically free cavity whose output is a complex combination of both mechanical and acousto-electrical effects.

The resonator's layout consists of identical input and output IDTs with an acoustic aperture of 800 μm and 2.4 μm finger width. The free cavity is 192 μm wide and is flanked on each side by 400 positive and negative reflector strips of 2.4 μm pitch.

The resonator layout was transferred via contact lithography onto a 36°-rotated Y-cut X-propagating lithium tantalate ($LiTaO_3$) piezoelectric substrate. The $LiTaO_3$ substrate possesses a high electromechanical coupling coefficient, thus rendering the SH-SAW sensors a high sensitivity to the electrical properties of the liquids. A photograph of the fabricated SH-SAW resonator sensor in shown in Figure 1.

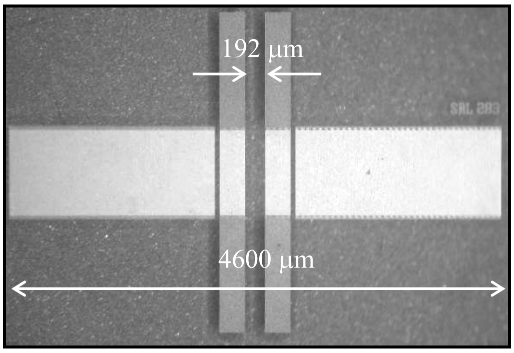

Figure 1: SH-SAW two port resonator layout

The two-port resonator sensor was used in conjunction with a micro-stereolithographically (MSL) made liquid reservoir placed on top of the sensor to host the test liquid. Electrical measurements were performed with a network analyser (Agilent 8753ES) from which the frequency, amplitude and phase information were recorded for each measurement. The resonator's amplitude and phase responses were used to differentiate between different liquid samples.

Amplitude and phase responses can be expressed as complex functions of the physical properties of the liquid, e.g. density, viscosity, conductivity and permittivity. The resonator sensor was previously able to discriminate between six basic taste solutions [7] and further investigations are now carried out on taste samples with different concentrations. Acetic acid (soured taste), sucrose (sweet taste) and sodium chloride (salty taste) solutions with 0.5%, 1%, 3% and 5% concentrations were tested to the resonator and the results confirm sensors ability to discriminate between these samples. The sensor response was assessed relative to the properties of the test solutions.

MULTIVARIATE ANALYSIS

Multiple linear regression (MLR) is applicable to situations where several independent variables (predictors) account for the variance of a dependent variable. The general formula describing the linear model is

$$y_i = b_0 + \sum_{j=1}^{k} b_j x_j + e \tag{1}$$

where y_i is the observation variable, also known as response or dependent variable and x_j are the independent variables (also called predictors, regressors or explanatory variables). The b_0 constant is the intercept point and b_j are the regression coefficients (sensitivities) for the corresponding x_j variables; each regression coefficient represents the average amount the dependent increases when the corresponding independent increases one unit and other independents are held constant. The error term or residual is denoted by *e*. Regression analysis employs linear algebra to perform the necessary calculations for solving equation (1).

Among the different criteria that can be used to best-fit a data-set, the least squares criterion is one of the most powerful. The least squares solution is computed by minimizing the sum of squares of the distances (deviations) from each observation point to the best-fit plane.

The model residuals are defined as the differences between the observed and predicted values. Residuals are useful for detecting failures in the model assumptions, since they correspond to the errors in the model equation (1). By assumption, these errors have independent normal distributions with mean 0 and constant variance. The smaller the variability of the residuals around the regression line relative to the overall variability, the better the model is.

To assess the goodness of fit, multiple linear regression uses the R^2 coefficient of multiple determination. Also known as the multiple correlation or Multiple-R coefficient, R^2 gives the correlation between the observed values and the values predicted by the regression model. The R^2 multiple coefficient represents the percent of the variance in the observed value explained jointly by the independent variables. It can also be interpreted as the proportionate reduction in error in estimating the dependent when knowing the independents.

The F statistic, also used as F ratio, is used to test the significance of R^2 and implicitly, the significance of the regression model as a whole. It provides information whether the regression model has improved the accuracy of predictions to a statistically significant level. F is a function of R^2, the number of independents (*k*) and the number of cases (*n*) and is computed with k and (*n*-*k*-1) degrees of freedom:

$$F = \frac{R^2 / k}{(1-R^2)/(n-k-1)} \tag{2}$$

If the p-value or prob(F) < 0.05 then the model is considered statistical significant at the 95% level and the null hypothesis of no linear relationship of the dependent to the independents (all regression coefficients are zero) can be rejected.

RESULTS AND DISCUSSIONS

Acetic acid, sucrose and sodium chloride measured data are plotted against the weight per cent concentration of each solution in Figures 2 and 3. A preliminary analysis of the amplitude and phase variations with concentration shows tendencies of quasi-linear dependencies. Regression plots (linear) have been added to the measured data to illustrate amplitude and phase evolution with increasing solute concentrations.

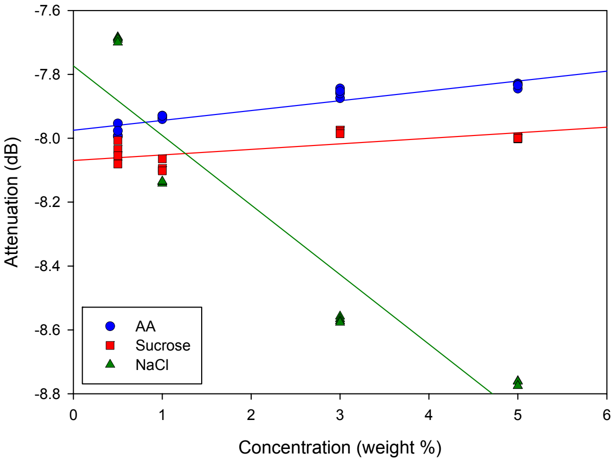

Figure 2: Amplitude variation with concentration

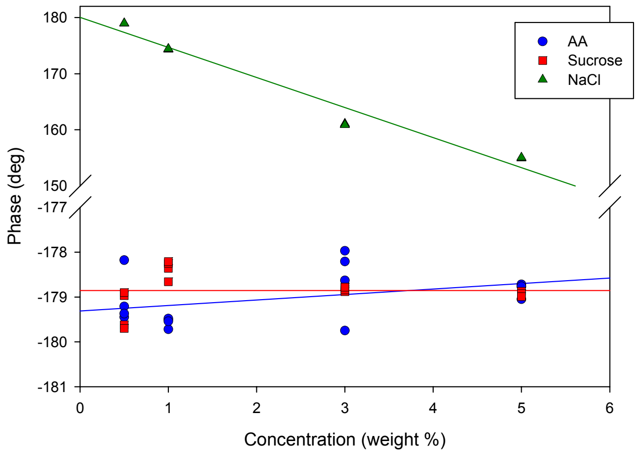

Figure 3: Phase variation with concentration

A multivariate analysis was then performed on the SH-SAW sensor's amplitude and phase data. Density, conductivity and permittivity were chosen as predictors for the multiple linear regression models. The purpose of the multivariate analysis was to verify whether the amplitude and phase data can be related to the selected physical parameters of the liquids and whether substance-specific multiple linear models are able to approximate the resonator's response with sufficient accuracy.

Multivariate analysis can be used to formulate tailored models for the resonator sensor response to various liquid samples, can help in identifying the dominant interactions that take place at the sensor-liquid interface and establish the relative magnitude of their effect.

Multiple linear regression models with high predictive power were obtained for the basic taste considered here: sourness, sweetness and saltiness. Figure 4 presents a typical scatter plot of the measured data together with the estimated values for acetic acid solutions (◊ denotes least squares estimates and + denotes robust fit estimates). Similarly, Figures 5 and 6 represent sucrose and NaCl measured and predicted data. A measure of the fit of each model is given by the R^2 multiple regression coefficient. The lowest values were obtained for sucrose data (0.72 and 0.88) while the highest values were achieved for NaCl data for both amplitude and phase (0.99). The amplitude and phase multiple linear regression models for acetic acid, sucrose and NaCl data are summarized in Tables 1-3. The models were found to approximate the measured data well and explain high percentage of the data variance (up to 0.97 for amplitude and 0.90 for phase for acetic acid solutions). The F-tests and p-values for each model allow rejection of the null hypothesis and confirm the linear dependency between the sensor response and the considered predictors. Multiple linear models can be used to approximate the resonator's response, offering a simple prediction tool and a valid alternative to the non-linear analytical models.

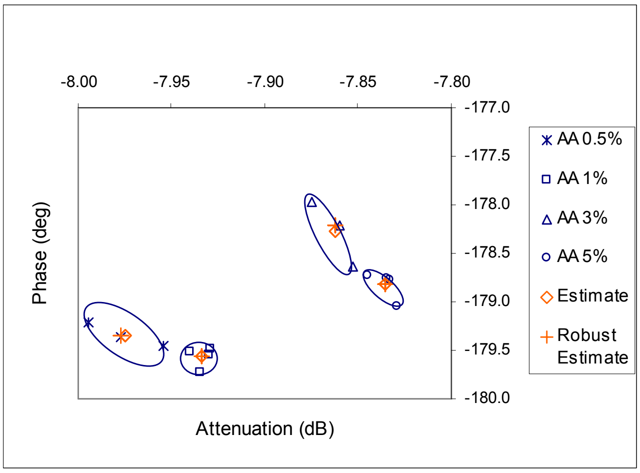

Figure 4: Acetic acid solutions with 0.5%, 1%, 3% and 5% concentrations and their predicted values

Table 1: Acetic acid regression models

Amplitude	Regression coefficients			
	b0	b1*ρ	b2* σ	b3*ε_r
	-97.224	0.077	1.353	0.144
	Model Statistics			
	R^2	F-test	p-value	error variance
	0.970	108.290	6.35E-08	0.0001
Phase	Regression coefficients			
	b0	b1*ρ	b2* σ	b3*ε_r
	-10638.1	9.091	-7.144	17.212
	Model Statistics			
	R^2	F-test	p-value	error variance
	0.903	31.102	2.21E-05	0.0356

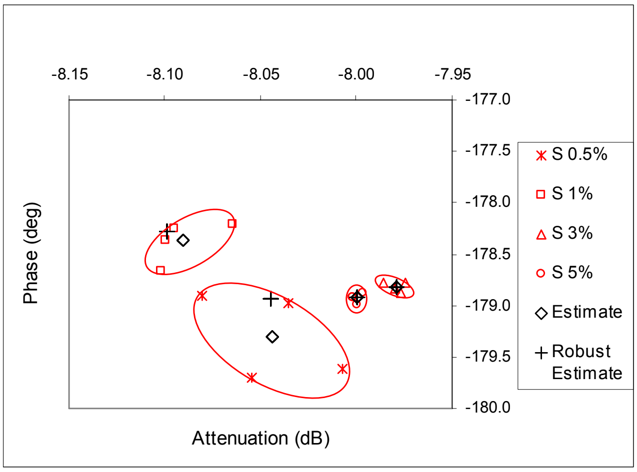

Figure 5: Sucrose solutions with 0.5%, 1%, 3% and 5% concentrations and their predicted values

Table 2: Sucrose regression models

	Regression coefficients			
Amplitude	b0	b1*ρ	b2* σ	b3*ε_r
	-3210.1	1.354	-2522.09	23.016
	Model Statistics			
	R^2	F-test	p-value	error variance
	0.882	30.041	0	0.0003
Phase	Regression coefficients			
	b0	b1*ρ	b2* σ	b3*ε_r
	13672.73	-5.880	-28875.4	-99.259
	Model Statistics			
	R^2	F-test	p-value	error variance
	0.723	10.439	0	0.0558

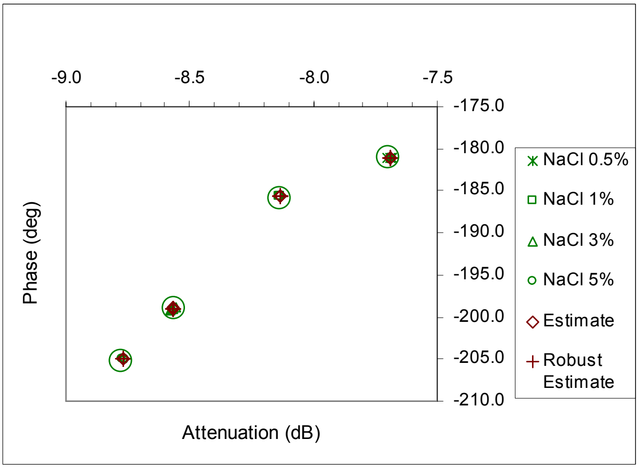

Figure 6: NaCl solutions with 0.5%, 1%, 3% and 5% concentrations and their predicted values

Table 3: NaCl regression models

	Regression coefficients			
Amplitude	b0	b1*ρ	b2* σ	b3*ε_r
	-301.782	0.390	-4.319	-1.186
	Model Statistics			
	R^2	F-test	p-value	error variance
	0.9997	17996.15	0	5.1E-05
Phase	Regression coefficients			
	b0	b1*ρ	b2* σ	b3*ε_r
	-5339.44	4.313	-7.353	10.649
	Model Statistics			
	R^2	F-test	p-value	error variance
	0.9999	100036.8	0	0.0050

CONCLUSIONS

A multivariate analysis has been carried out on the response of a 433 MHz SAW resonator (amplitude and phase data) with the liquid parameters of density, conductivity and electrical permittivity used as predictors. Multiple linear regression models with high predictive power were obtained for all the basic taste solutions considered here. To the authors' knowledge, this is the first attempt to map the physical properties of liquid samples to the taste characteristics for a SH-SAW liquid sensor. The derived multivariate models can be used with high confidence to approximate the resonator's response, offering thus a simple prediction tool and a valid alternative to the non-linear analytical models. We recognize that this (like electrochemical cells) is an indirect measurement of taste but could nonetheless be a useful analytical tool.

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