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Design of Electronic Nose System Using Gas Chromatography Principle and Surface Acoustic Wave Sensor

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Abstract

Most gases are odorless, colorless and also hazard to be sensed by the human olfactory system. Hence, an electronic nose system is required for the gas classification process. This study presents the design of electronic nose system using a combination of Gas Chromatography Column and a Surface Acoustic Wave (SAW). The Gas Chromatography Column is a technique based on the compound partition at a certain temperature. Whereas, the SAW sensor works based on the resonant frequency change. In this study, gas samples including methanol, acetonitrile, and benzene are used for system performance measurement. Each gas sample generates a specific acoustic signal data in the form of a frequency change recorded by the SAW sensor. Then, the acoustic signal data is analyzed to obtain the acoustic features, i.e. the peak amplitude, the negative slope, the positive slope, and the length. The Support Vector Machine (SVM) method using the acoustic feature as its input parameters are applied to classify the gas sample. Radial Basis Function is used to build the optimal hyperplane model which devided into two processes i.e., the training process and the external validation process. According to the result performance, the training process has the accuracy of 98.7% and the external validation process has the accuracy of 93.3%. Our electronic nose system has the average sensitivity of 51.43 Hz/mL to sense the gas samples.

Keywords*: acoustic features, gas chromatography (GC), hyperplane, support vector machine (SVM), surface acoustic wave (SAW)*

1. Introduction

Gas is a matter which has an independent shape but tends to expand indefinitely. Most gases are colorless and odorless which difficult to be sensed by the naked eye and human olfactory system. In addition, the gases which result in toxic odor are forbidden to be sensed using the human nose directly [1]. Therefore, an electronic device is required for gas recognition. Over the last decades, the electronic nose device has extensively been used in industry for the quality monitoring system, gas identification, chemical analysis, etc. Electronic nose technology refers to the capability of the human olfaction using a sensor configuration and a pattern recognition algorithm [2,3].

In the electronic nose system, a sensor array is required to sense the odor. The Metal-Oxide-Semiconductor (MOS) sensor such as Taguchi Gas Sensor (TGS) becomes the type of sensor widely used for gas sensing applications due to its simplicity [4,5]. However, it has the low sensitivity which generally requires the high sample of concentration, i.e., in the range of parts per million (ppm) level [6]. Another common gas sensor is quartz crystal microbalance (QCM), which is able to sense the odor at very low concentrations, i.e., single parts per million (ppm) or even parts per billion (ppb) [7,8]. To obtain a sensitive gas sensing, an array of QCM sensors is used in the electronic nose [9,10]. However, the main problems of these sensors can lead to complexity and interference. Therefore, in this study, we constructed the electronic nose system which has the simple configuration with high sensitivity and good repeatability. A Surface Acoustic Wave (SAW) sensor was selected as the detector. Principally, both SAW

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and QCM work based on the resonant frequency change. In the analytical approximation, Sauerbrey's formula presented in Equation 1 is widely used to determine the change of resonant frequency affected by the absorbed mass on the crystal's surfaces [11]:

$$\Delta F = -\frac{2F_0^{\ 2}}{A\sqrt{\rho\mu}}\Delta m \tag{1}$$

where ΔF is the change of resonant frequency (Hz), F_0 is the resonant frequency (Hz), Δm is the mass change (g), A is the active crystal area (cm^2), ρ is the crystal density (g/cm^3), and μ is the shear modulus of the crystal (g/cms^2).

In 2014, Hari Agus Sujono *et al.,* applied QCM sensor arrays for vapor identification system which has the resonant frequency of 20 MHz [9]. This type of sensor array induces the complex configuration and the interference issues. Therefore, only a single sensor of SAW will be used to sense the odor which has the resonant frequency of 34 MHz. The SAW sensor used in this experiment operates at a higher resonant frequency. Hence it affects the increase in sensitivity because the change of the resonant frequency (ΔF) to sense the mass change absorbed by crystal area for both sensors are dependent on their resonant frequency (F_0) as explained by Sauerbrey's formula.

To achieve a good selectivity in the electronic nose system, we applied a Gas Chromatography (GC) principle for the compound analysis. The GC is a technique based on the compound partition at a certain temperature which involves the two phases, i.e., the stationary phase and the mobile gas phase. The stationary phase material is located in the chromatography column as the partition material, whereas the mobile gas phase consists of a sample carried by dry air into the partition column [12]. Each sample has different elution strength because of the polarity suitability with the stationary phase material in the partition column [13]. In 2016, the electronic nose system by integrating GC and TGS sensor was conducted by Radi et al. [12]. However, the TGS sensor has a low sensitivity which requires the high amount of concentration for the measurement. Therefore, in this study, a combination between GC and SAW sensor in the electronic system is expected to overcome the issues.

For the recognition part in the electronic system, we used a learning algorithm of Support Vector Machine (SVM) for the classification process. The SVM is proposed as an effective technique for data classification. It is derived from statistical learning theory introduced by Vladimir Vapnik et al. [14]. Basically, another competitive learning algorithm is Artificial Neural Network (ANN), both of them are included in the supervised learning classifier [15]. However, many researchers reported that SVM classifier often outperforms than the ANN classifier [16]. The ANN classifier achieves an optimal local solution, while the SVM classifier obtains an optimal global solution. It is not surprising that the solution of the ANN classifier is different for each training process which results in a different optimal solution, whereas the solution offered by SVM classifier is same for every running. Hence, it generates the same optimal solution [17-19]. The contents of this paper are organized as follows: section 2 discusses the experimental design of the electronic nose system, feature extraction, and elaborate the SVM classifier technique. Furthermore, section 3 demonstrates the results and verification analysis. Finally, we present our conclusion in section 4.

2. Research Method
2.1. The Experimental Design

In this study, the experimental design of the electronic nose system includes four main parts, i.e., a gas sample, a GC column, a detector, and data analysis. Three types of gas samples were used in this study, i.e., methanol, acetonitrile, and benzene. The chromatography column consisted of Thermon-3000 and ShinCarbon as the stationary phase material. The SAW device which has the resonant frequency of 34 MHz was used as the detector to record the frequency change of the acoustic signal generated by each gas sample.

2.2. The Experimental Procedure

Figure 1 presents the design of our electronic nose system. The experimental setup is depicted in Figure 1a, whereas the schematic layout is shown in Figure 1b. There are three main parts of our design, i.e. the carrier gas, the chromatography column, and the SAW sensor. The dry air is used to carry the gas samples of A (methanol), B (acetonitrile), and C (benzene). The chromatography column is made of the glass material with the diameter of 3.2 mm and the length of 1.6 m. It has the operating temperature value above 70°C. The chamber as the oven wherein the chromatography column located is made of the aluminum with the geometrical size of 40 cmx7.5 cmx8 cm.

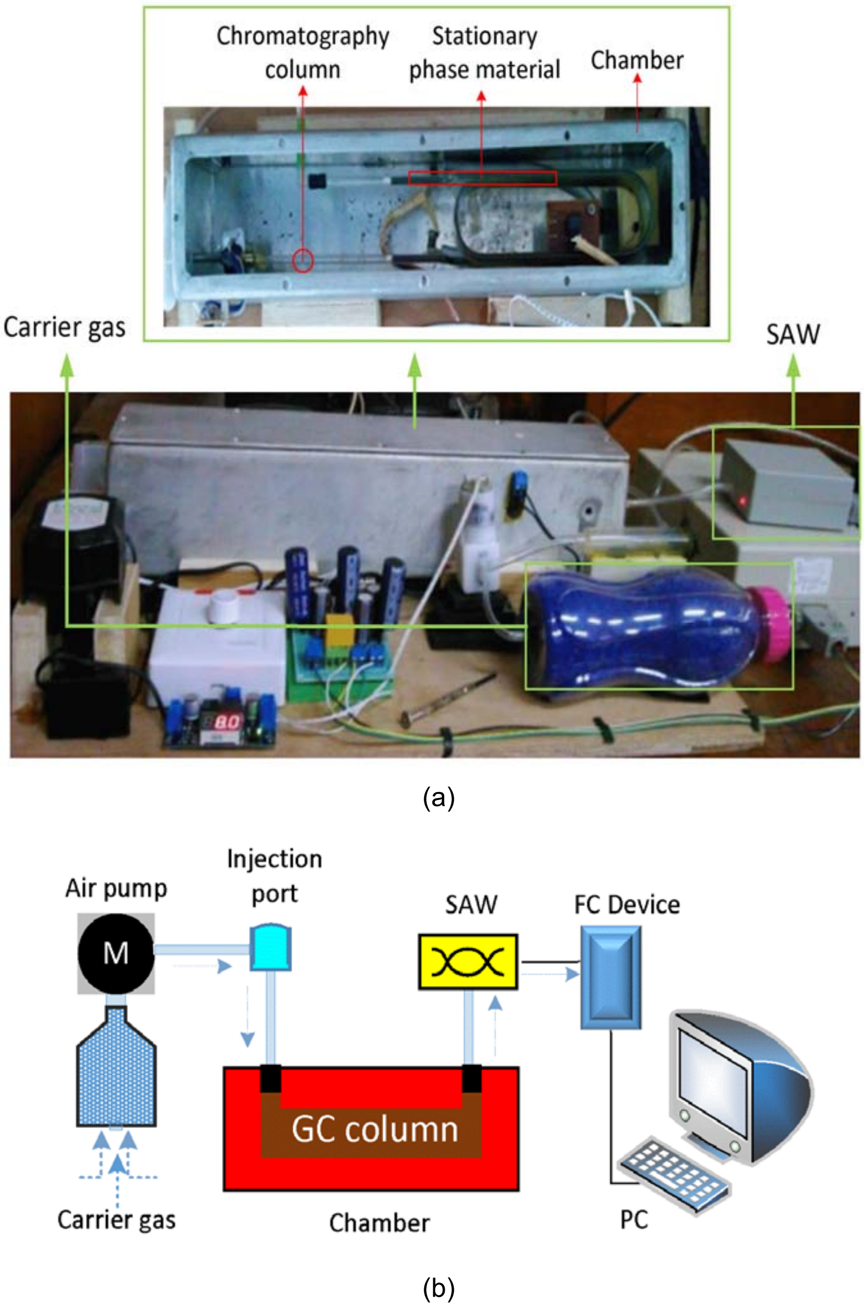


Figure 1. The design of electronic nose system: (a) the experimental setup, (b) the schematic layout

Before starting the measurement, the chromatography column is needed to be cleaned for 30 minutes using the carrier gas pushed by the air pump. The amounts odor volumes of 20 mL gas sample are introduced into the injection port. Then the gas sample is transported by the carrier gas into the chromatography column which is located in a chamber operated under the controlled temperature of 80^{0}C. Interactions between stationary phase material and the gas sample compound generate a series of fractions which is converted by the SAW sensor as the acoustic frequency change data. Furthermore, the acoustic signal data are transmitted to the computer through the Frequency Counter (FC) device for the data analysis. According to the measurement, the SAW sensor records the frequency value of about 34 MHz at initial condition before injecting the gas sample. The frequency change is described as

$$\Delta f = f(t) - f_{ref}. \qquad (2)$$

where Δf is the frequency change, f_{ref} is the initial frequency of 34 MHz, and $f(t)$ is detected frequency after injecting the gas sample. Furthermore, to collect the acoustic signal data produced by each gas sample needs 500 seconds. Figure 2 shows the sensor response to the acetonitrile.

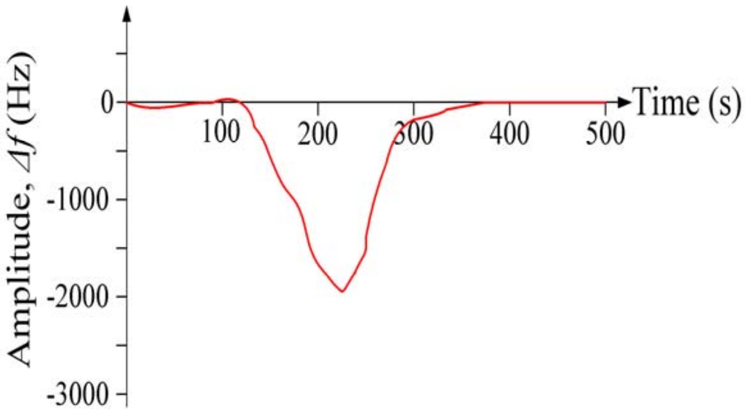


Figure 2. The sensor response to the gas sample of acetonitrile

2.3. Feature Extraction of Acoustic Signal Processing

In this study, the acoustic signal data would be processed to obtain the acoustic features. Figure 3 describes the parameters included to determine the acoustic features using the threshold of -100 Hz value. The four acoustic features including the peak amplitude A_p, the negative slope $S^{(-)}$, the positive slope $S^{(+)}$, and the length L are used in this study determined in Equation 3,4,5 and 6 respectively:

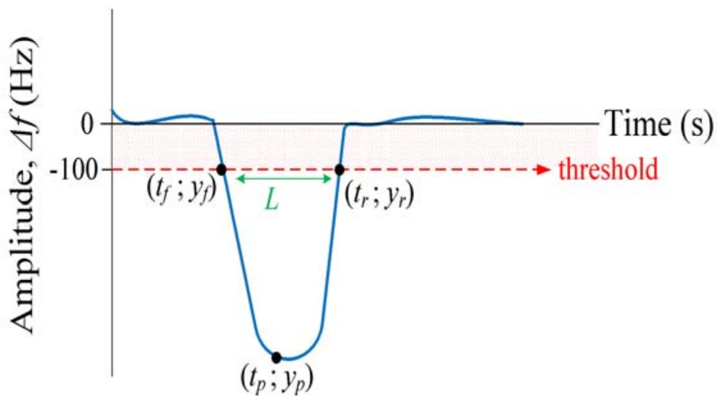


Figure 3. Acoustic feature parameters

$$A_p = y_p \tag{3}$$

$$S^{(-)} = \frac{y_p - y_f}{t_p - t_r} \tag{4}$$

$$S^{(+)} = \frac{y_r - y_p}{t_r - t_p} \tag{5}$$

$$L = t_r - t_f \tag{6}$$

where t_f is the fall time, y_f is the fall amplitude, t_p is the peak time, y_p is the peak amplitude, t_r is the rise time, and y_r is the rise amplitude.

2.4. Support Vector Machine (SVM) Classifier

In this study, we used the SVM classifier to identify the gas type which included four acoustic features as the input parameters. The gas types are divided into three classes. To understand the basic principle of SVM classifier, the simple linear separable case is shown in Figure 4. In the original space, a linear hyperplane $f(x)$ is used to separate the data points according to the support vector position. Hence, the linear hyperplane $f(x)$ groups the data points into two classes, i.e., class +1 and class -1 which are constrained by $f(x) \geq +1$ and $f(x) \leq +1$, respectively [20]. However, many real cases contain noise or outlier data points which are non-linearly separable [21]. Thus, the main objective of the SVM classifier is to obtain the optimal hyperplane model that can maximize the margin (M) of the classes [22].

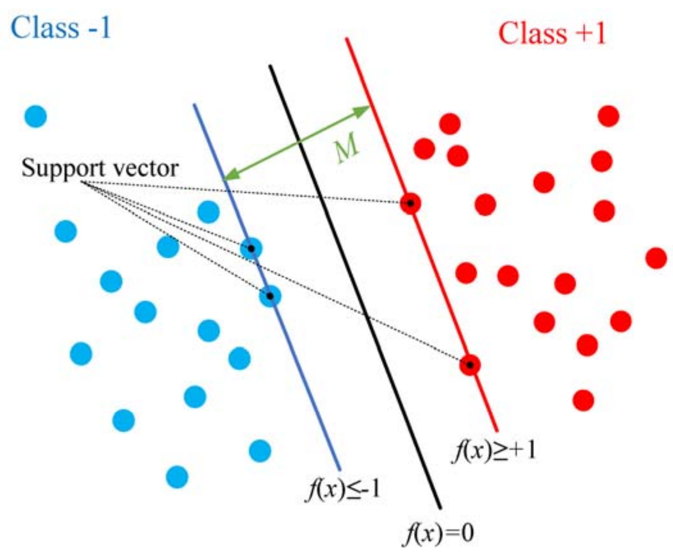


Figure 4. The Linear separable case using SVM method

The SVM classifier includes the kernels to optimize the hyperplane model for a nonlinear separable case. The kernels allow transforming the data points into a higher dimensional space called feature space to obtain a linear hyperplane although occasionally result in a nonlinear hyperplane in the original space. In this study, we used Radial Basis Function (RBF) as the kernel function. The RBF kernel is derived in Equation 7. Then the hyperplane model $f(x_d)$ is determined in Equation 8 [23,24].

$$\boldsymbol{H}(\boldsymbol{x}_i, \boldsymbol{x}_d) = \exp(-\gamma \|\boldsymbol{x}_i - \boldsymbol{x}_d\|^2) \tag{7}$$

$$f(\boldsymbol{x}_d) = \sum_{i=1}^{n} \alpha_i y_i \boldsymbol{H}(\boldsymbol{x}_i, \boldsymbol{x}_d) + b \tag{8}$$

where x_d is the data point, α is lagrange multiplier, y_i is membership of the gas sample class, γ is gamma, x_i is support vector, b is intercept, and i = 1, 2, 3, . . . , n.

It is broadly mentioned that the SVM classifier using RBF kernel requires the best combination of two hyperparameters, i.e., gamma (γ) and cost (c) to build the optimal hyperplane model. The gamma explains how significant the influence of each data point in the training set. For example, a higher value of gamma leads the over-fitting problem because it tries to fit exactly each data point in the training set. Whereas, the cost is used to control the trade-off between smooth decision boundary and classifying the data points in the training set correctly [25,26].

In this study, the identification gas system using SVM classifier consisted of two processes, i.e. the training process and the external validation process. The training process was used to build the hyperplane model which included the acoustic signal data from the total numbers of 150 gas samples. Since the external validation process was used to assess the SVM performance. It used the acoustic signal data obtained from the total numbers of 30 gas sample measurements.

To describe the performance analysis of the SVM classifier, the 3x3 confusion matrix table was applied for this study, shown in Table 1 [27]. In the confusion matrix table, the actual result is the data based on the observation (reality). It is consisted of three classes i.e., class A, B, and C, whereas the predicted result is the identification result assessed by the SVM classifier which also consisted of three classes. The cases are divided into nine values: *TA*, *FA1*, *FA2*, *FB1*, *TB*, *FB2*, *FC1*, *FC2*, and *TC*. Finally, the accuracy (*AC*) used to assess the SVM performance to classify gas sample is determined in Equation 9 [28].

Table 1. The 3x3 confusion matrix

		Actual Result		
		A	B	C
Predicted Result	A	*TA*	*FA1*	*FA2*
	B	*FB1*	*TB*	*FB2*
	C	*FC1*	*FC2*	*TC*

$$AC = \frac{T}{T + FA1 + FA2 + FB1 + FB2 + FC1 + FC2} \times 100\% \tag{9}$$

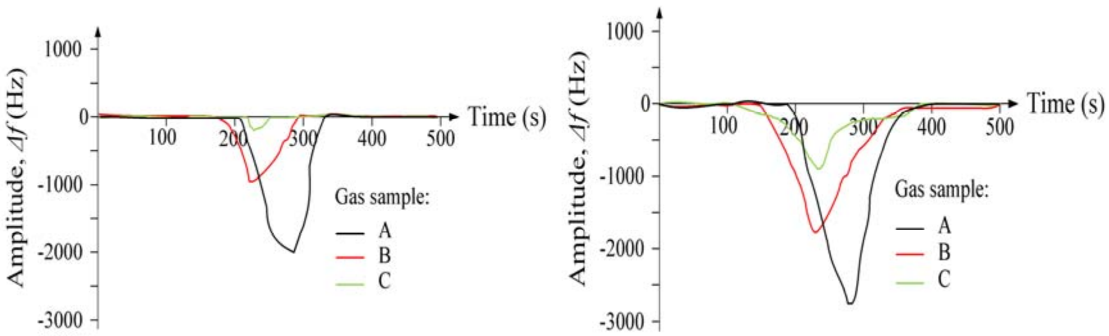
where $T = TA + TB + TC$,*TA* is the correctly classified class A, *TB* is the correctly classified class B, *TC* is the correctly classified class C, *FA1* is the class B classified into class A, *FA2* is the class C classified into class A, *FB1* is the class A classified into class B, *FB2* is the class C classified into class B, *FC1* is the class A classified into class C, *FC2* is the class B classified into class C.

3. Results and Analysis

In this study, we designed the electronic nose system which concerns into two terms, i.e., having high sensitivity and good repeatability. Figure 5 shows the measurement result of the signal acoustic data recorded by the electronic nose system. Based on the Equation 2, each gas sample has the specific frequency change (Δf) curve affected by each gas sample's mass absorbed by crystal area of SAW sensors. In the term of sensitivity, according to Figure 5a by using 5 mL odor volume, the amplitude peak determined in equation 3 from gas sample A, B, and C are -2000 Hz, -1000 Hz, and -85 Hz, respectively.

Furthermore, by applying 20 mL odor volumes, the gas sample A, B, and C reach the amplitude peak of -2800 Hz, -1750 Hz, and -850 Hz, respectively. It means that the electronic nose system could sense the odor from methanol, acetonitrile, and benzene with the sensitivity of 53.3 Hz/mL, 50 Hz/mL, and 51 Hz/mL, respectively. Hence the average sensitivity is 51.43

Hz/mL. Rivai et al. [29,30] have designed the electronic nose using gas chromatography and QCM sensor. According to the result performance, it has the approximate sensitivity of 6.5 Hz/mL to sense the odor of ethanol. Another research paper shows that the frequency change curve resulted by specific odor, fragrance, and gas in the range of hundreds herzt [31]. Our system offers higher operating resonant frequency. Hence, it is able to generate the specific or distinctive acoustic signal coming from each gas sample in the range of thousands hertz, shown in Figure 5.

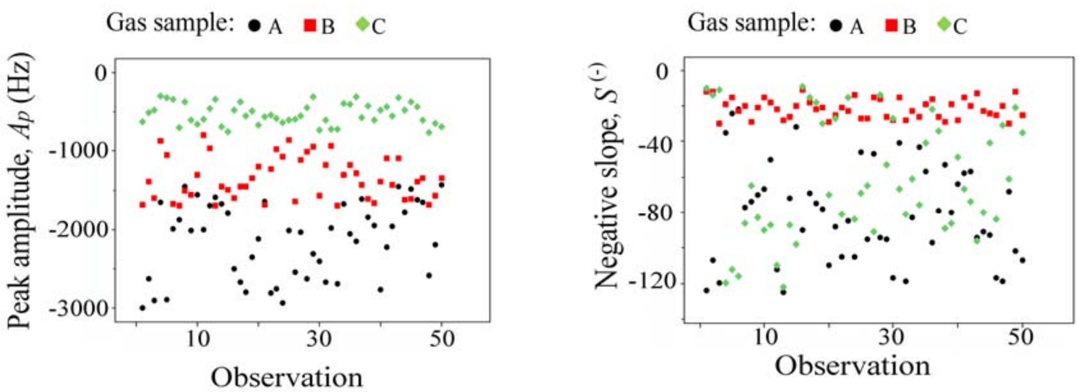


(a) (b)

Figure 5. The acoustic signal data affected by the odor volume of (a) 5 mL, and (b) 20 mL

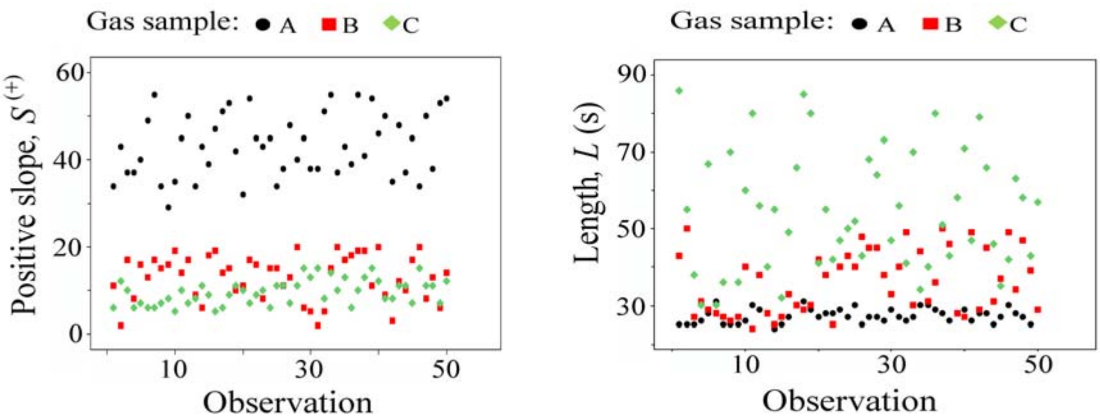
In the term of repeatability, each gas sample compound produces a specific acoustic signal data because it has particular interactions with stationary phase material in the chromatography column. The main difference of each gas sample curve is pointed out by its peak amplitude A_p. For example, in Figure 5a, the highest peak amplitude is achieved by gas sample C. The gas sample A has the lowest value of amplitude peak and these trends are repeated in Figure 5b when using the amounts odor volumes of 20 mL. Furthermore, the acoustic signal data generated by each gas sample are processed to obtain the four acoustic features.

Figure 6 presents the distribution of four acoustic features i.e., the peak amplitude A_p, the negative slope $S^{(-)}$, the positive slope $S^{(+)}$, and the length L which refer to Equation 3, 4, 5, and 6 respectively. The distribution result given by Figure 6 includes 50 measurements using 20 mL odor volumes. The distribution results of the peak amplitude A_p, the negative slope $S^{(-)}$, the positive slope $S^{(+)}$, and the length L can be seen in Figure 6a, 6b, 6c, and 6d sequentially. As shown in Figure 6a, 6b, 6c, and 6d, we can conclude that the distribution of the acoustic features from gas sample A, B, and C are classified into the non-linearly separable case.



(a) (b)

Figure 6. The acoustic features of gas sample: (a) the peak amplitude, (b) the negative slope.



(c) (d)

Figure 6. The acoustic features of gas sample: (c) the positive slope, (d) the length

The recognition algorithm of SVM classifier was used to solve the non-linearly separable case. In this study, the total numbers of 150 gas samples were used for training process to build the optimal hyperplane model using RBF kernel. The RBF kernel requires the best combination of two hyperparameters of gamma and cost. In the hyperparameters tuning, we set the interval of gamma started from 2^{-15} to 2^{2}, whereas the cost has the lower limit of 2^{-15} and the upper limit of 0.25. Figure 7 presents the detail distribution of the hyperparameter tuning performance. The dark blue color (left) and the dark green color (right) have the lowest and highest accuracy, respectively. According to the training process, the best combinations of gamma and cost are 1 and 0.2, respectively, which have the accuracy of 98.7%. It means the total numbers of 150 observations used for the training process: 148 data are correctly classified and the others are incorrectly classified.

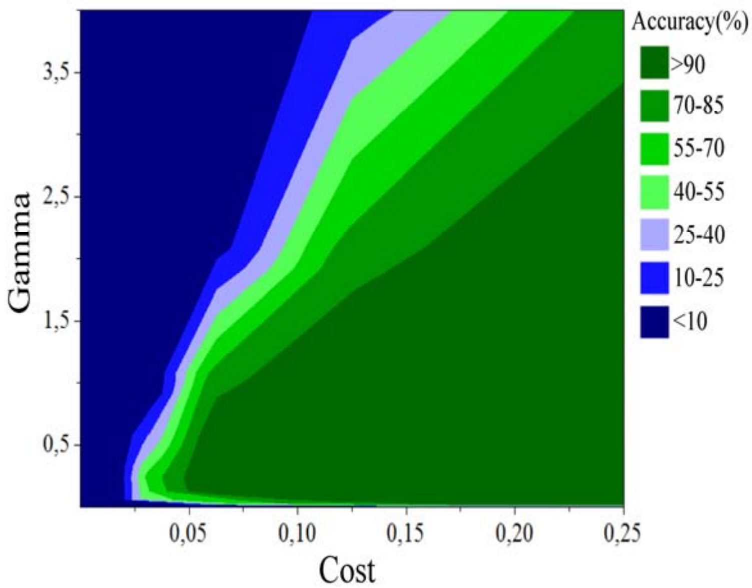


Figure 7. The hyperparameters tuning performance

The external validation was used for the testing to assess the SVM classifier performance. It included the acoustic signal data from 30 gas samples (10 data for each gas sample). Table 2 shows the confusion matrix result from external validation process. The result of case *TA*, *FA1*, *FA2*, *FB1*, *TB*, *FB2*, *FC1*, *FC2*, and *TC* are 9, 1, 0, 1, 9, 0, 0, 0, and 10, respectively. The SVM classifier has the accuracy of 93.3% to identify the gas sample, which means from the total numbers of 30 observations used for the external validation process: 28 correctly classified and 2 incorrectly classified. These results indicate that the SVM classifier can be categorized as the robust algorithm which can be integrated with the electronic nose system.

Table 2. The 3x3 confusion matrix result

		Actual Result		
		A	B	C
Predicted Result	A	9	1	0
	B	1	9	0
	C	0	0	10

4. Conclusion

The design of the electronic nose system with good repeatability and high sensitivity by integrating the gas chromatography principle and Surface Acoustic Wave (SAW) sensor was successfully demonstrated. Three gas samples were used for the measurement process, i.e., methanol, acetonitrile, and benzene. In the previous research, the electronic nose using gas chromatography and QCM sensor has only the approximate sensitivity of 6.5 Hz/mL to sense the odor. Another research also shows that the frequency change curve resulted by specific odor, fragrance, and gas only in the range of hundreds herzt. Based on the result analysis, our electronic nose system has the average sensitivity of 51.43 Hz/mL and also offers higher operating resonant frequency. Hence, it is able to generate more specific or distinctive acoustic signal coming from each gas sample.

The repeatability performance is shown by the distinctive acoustic signal curve from each gas sample due to the specific interactions between the odor and the material located in the chromatography column. In this study, Support Vector Machine using Radial Basis Function was applied to recognize the odor. The four acoustic features obtained from acoustic signal data were used for input parameters in the classifier, i.e., the amplitude peak, the negative slope, the positive slope, and the length. The classification using Support Vector Machine was divided into two processes, i.e., the training process and the external validation process. The training process and the external validation process have the high accuracy of 98.7% and 93.3%, respectively. These results indicate that the classifier can be applied to the electronic nose system. Finally, to achieve the comprehensive result performance, the future work will concern on a deep investigation of sensitivity and repeatability performances at the electronic nose system by varying the temperature of the chamber, the pressure of the air pump, and different type kernel functions in the Support Vector Machine algorithm.

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