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ON-LINE MONITORING OF THEAFLAVINS AND THEARUBIGINS RATIO IN TURKISH BLACK TEA USING ELECTRONIC NOSE

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ABSTRACT

Black tea is one of the most popular beverages all over the world. Although there are a lot of tea types, black tea has the highest consumption ratio. The production sequence of black tea begins with 45 minutes of a curling process for disintegrating fresh tea leaves after 16-20 hours of withering, it continues with 135 minutes of fermentation within a 27°C, 95% humidity and finishes with the drying process. Fermentation is accepted as the most crucial step of the overall process, either under-fermentation or over-fermentation leads to deterioration of the finished tea quality. Quality test of the finished tea is made by both tea testers and chemical analysis. Many researches have been performed for testing the quality of the finished tea and classifying it electronically by taking tea samples to a laboratory environment which results with high classification performances. In addition to previous studies, this study performed an online electronic measurement on the fermentation line of a tea production facility.

In this study, a sensor block with 13 metal oxide semi-conductor (MOS) sensors has been built and measurements have been made. The sniffed tea has also been taken to laboratory for chemical analysis (theaflavins (tf), thearubigins (tr)) to determine the quality of tea. After determining tf/tr ratio, the tea was classified into three subgroups as low, mid and high quality. 5 sensors which are related with the tea odour were detected. Five features were extracted from 5 sensors' data and then, classifications have been made by using Linear Discriminant Analysis (LDA), Bayes and k-Nearest Neighbour (k-NN) algorithms. The proposed method was successfully applied to the data sets and achieved a classification rate of 74.19% on the test data by using the k-NN-3. Quality analysis of Turkish Black Tea was done by electronic nose.

Key Words: black tea quality, electronic nose, gas sensors, k-NN method.

1. INTRODUCTION

Tea is a kind of beverage which has the second highest consumption ratio after water and its drinking habit is increasing day by day all over the world [1]. Tea types and consumption habits are highly variable. For instance, while western countries mostly consume black tea, eastern countries prefer green tea [2]. According to different resources with little variations, black tea has the highest production ratio over all tea types by 70%, green tea and oolong tea with others have 23% and 7% respectively [3]. During the production sequence, if the tea is dried after collecting, it becomes green tea; if it is dried after half fermentation, it becomes oolong tea or if it is dried after full fermentation, it becomes black tea.

Black tea production sequence includes; withering, curling, fermentation and drying process respectively. Fermentation begins with the curling process and it lasts until the drying process. Tea production facilities in Turkey usually prefer 3 hours of fermentation including the curling state. Tea reaches the drying state after around 45 minutes of curling and 135 minutes of the fermentation line. Without knowing the quality, tea is being classified by tea tester experts according to its taste, colour and odour. After classification, tea passes through the blending process and packaging line to prepare for marketing. Although all these processes have effect, the fermentation is the most effecting state in the final quality of tea [4].

Black tea's quality could also be determined by chemical analysis methods by measuring and calculating the ratio of tf and tr levels in the tea. Tf/tr ratio is accepted as 1/10 for a high quality tea while mid quality tea has a ratio between 1/25 – 1/20 [4]. Although tf/tr ratio of tea which precisely determines tea's colour, odour, hardness and aroma could be calculated by chemical analysis, this method is both expensive and time consuming. Tea experts' evaluation method also has both relativity and cost disadvantages. Different experts could give different results and also the results are directly related to expert's health status. With this kind of studies it would be possible to determine the quality of tea precisely at any time without any relativity.

There are many researches on this topic to extract quality information from tea by using an electronic nose (e-nose), image processing (e-vision) and an electronic tongue (e-tongue). Irina Yaroshenko and her friends conducted a tea quality determination experiment and they studied the relation between tea tester experts' result and the chemical analysis results which was gathered with an e-tongue from finished tea liquor by brewing tea with a certain amount of boiling water for 5 minutes [5]. Mousumi Palit and his friends studied black tea which was first classified by experts then they used an e-tongue for collecting data from tea brewed with hot water. This data is used in both studies for classification as training and test data by 90% to 10% and 60% to 40% respectively and achieved accuracy over 90% [6,7]. With another experiment, Runu Banerjee (Roy), Nabarun Bhattacharyya and their friends analysed the faults of tea quality determination methods by collecting data using the e-tongue, e-nose and combinations with 40 grams of black tea brewed with hot water [8]. Another tea quality determination study with the e-tongue was carried out by Subrata Sarkar and his friends. They classified the black tea according to experts' points by brewing 2.5 grams tea with 180 ml boiling water for 5 minutes [9].

Image processing is another method and used in several studies to determine tea quality. Yuerong Liang and his friends observed the relation between the colour and quality of tea [10]. Mona Sharma and her friends used the image processing method to determine optimum fermentation duration and indicated the relation between the fermentation duration and the size of tea leaves fragments [11]. Amitava Akuli and his friends studied with image processing method to determine tf/tr level by brewing finished tea with boiling water for 5 minutes and achieved 77% classification accuracy [12].

The e-nose is also used to determine tea quality. Ritaban Dutta and his friends classified 5 different types of black tea using the e-nose include 4 sensors with 100% accuracy [13]. Nabarun Bhattacharyya, Bipan Tudu and their friends first used a commercial grade e-nose (Alpha MOS 2000) and classified 6 differently fermented finished orthodox black tea with 100% accuracy [14], then used their own e-nose with different gas sensor quantities and classified the brewed tea according to fermentation durations and quality with high accuracy [15-18].

Santi Sankar Chowdhuryl and his friends built a portable e-nose and classified the tea according to experts' points for two different tea facilities with Multilayer Perceptron (BP-MLP) and feed-forward Multilayer Perceptron (FFMLP) methods accuracy around 80,5% - 85,7% and 78,3% - 83,6% respectively [19]. Ashis Tripathy and his friends used several methods as Principal Component Analysis (PCA), Linear Discriminant Analysis (LDA), Kernel based Principal Component Analysis (KPCA) and Kernel based Linear Discriminant Analysis (KLDA) for tea quality classification and achieved accuracy of around 56.78%, 56.79%, 78.00% and 84.99% respectively [20].

Bipan Tudu and his friends placed 50 grams at the fermentation state black tea samples to a sample box then transferred the odour to an e-nose with 5 sensors to capture data. This data is used 90% as training and 10% as test, classified by fuzzy logic algorithm with 80% accuracy [21].

With these researches, scientists are able to determine the quality of black tea by using the e-nose, e-tongue and e-vision systems with high classification performances. Yet, all of these studies have been performed in a laboratory environment with a certain amount of pre-processed black tea which is either finished or in the fermentation state. Our study performed a black tea quality test without any pre-processing state, on the fermentation line with an on-line electronic nose during the fermentation state.

Following in the paper, chapter 2 explains the architecture of the e-nose built for this study, chapter 3 includes classification algorithms, chapter 4 shows experimental results, chapter 5 is the discussion, and chapter 6 is the conclusion.

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2. EXPERIMENTAL SETUP

A total 13 Figaro brand MOS sensors, given in Table-1, with 14 outputs are assembled to an 11x11 cm. printed circuit board (PCB) and inserted into a 13x14x13 cm. aluminium odour chamber with a 1 cm. wall. The sensor array and odour chamber can be seen in Figure 1-a and Figure 1-b.

Table 1. Sensors used in electronic nose.

TGS 813 –A00	Combustibele Gases, HC
TGS 825	Hydrogen Sulfide
TGS 826	Ammonia/Amine/Odour
TGS 830	Halocarbon Gases (CFC, HCFC)
TGS 880	Ethanol
TGS 2104	Air Quality/ Gasoline Exhaust
TGS 2180	Microwave Oven/ Water Vapour
TGS 2201	Air Quality/ Dual Sensor Element
TGS 2602	Air Quality/ Odour
TGS 2610-D00	Propane,Butane, LPG
TGS 2611-C00	Methane, Natural Gas
TGS 2620	Alcohol, Organic Vapour
TGS 5042	Carbon Monoxide (CO)

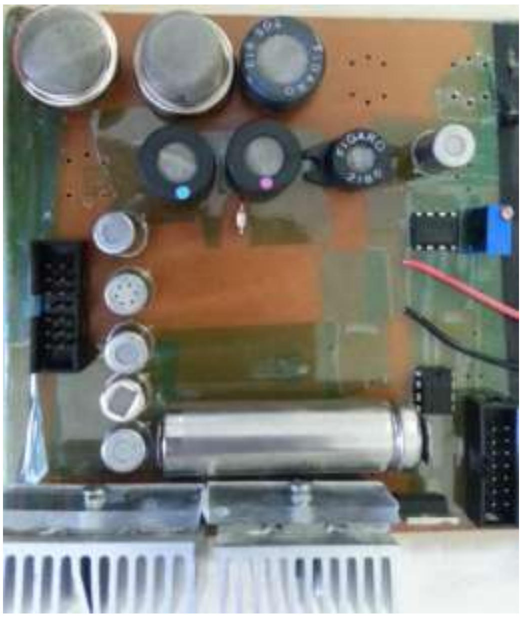

Figure 1-a. Sensor Array.

Figure 1-b. Odour chamber.

The odour chamber has one air input and one air output. The input side is separated into two parts, the first part is used for the vacuum pump that sucks the air from the fermentation line, and the second part is used for the oxygen tank to clean the sensor for a reference odour. The output side is connected to another vacuum pump to evacuate sniffed air. An odourless, teflon hose is used between the vacuum pumps and the chamber; also three solenoids are used to manage the air flow. Sensors' output values are sampled with two separate DAQ (National Instrument USB-6008 ve USB-6009) and stored to a computer via a MATLAB Simulink program. The complete system schematic of the built e-nose is shown in Figure 2.

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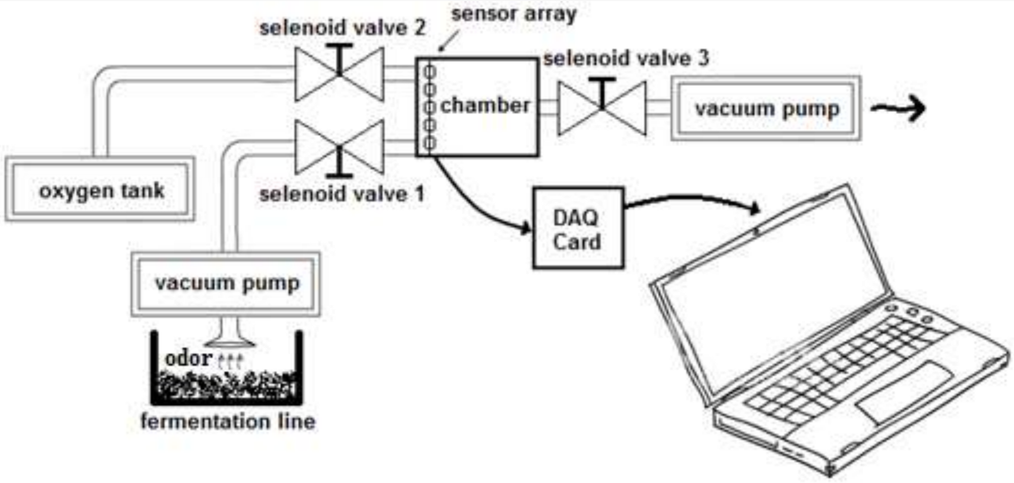

Figure 2. Odours capturing system.

One single sniffing cycle takes 60 seconds and has the following steps:

Step 1. Sensor Cleaning Process: Solenoids number 2 and 3 are opened first to clean the chamber and sensors to a reference odour for 10 seconds. This means for every sniffing, the process will have the same reference odour. Oxygen is chosen instead of outside air to clean the sensors because the facility has a strong tea fermentation scent.

Step 2. Sniffing Process: In the second step, solenoids number 1 and 3 are opened for 30 seconds to have continuous air flow to the sensors via air circulation in the chamber.

Step 3. Odour Lock Process: All solenoids are closed for this step to lock the odour in the chamber for 10 seconds to monitor sensors' output with non-circulating air in case of possibility of some sensors' slow reaction. This can be calculated as, 5 V applied to TGS-2602 by connecting a serial 470Ω and sensor's conductivity is calculated as in the Equation (1).

$$\rho = \frac{1}{R_S} = \frac{I_S}{V_S} = \frac{\frac{V_L}{470}}{5 - V_L} \qquad (1)$$

Here R_s, I_s, V_s and V_L represent sensor resistance, current, sensor supply voltage and serial resistance voltage respectively. The conductivity of the sensor changes with gas density, so this enables us to observe changing of the gas concentration by following voltage on this resistance.

The fermentation line has 1.8 m. width, 30 m. length and spread out tea has a 25 cm height.

Fermentation environment has the following specifications:

Humidity: 90%-95%

Temperature: 27-28^{o}C

Ambient Temperature: 24-25^{o}C

Odour measurements were taken from facility's fermentation line by the portable e-nose, shown in Figure 3-a and Figure 3-b, at 64 different locations around the centre of the line and 10 cm above the tea level. Two hundred grams of tea was taken from the measured points and dried in a stove to fix the quality and aroma to be the same at the sniffing moment.

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Figure 3-a. Online sniffing process over the fermentation line.

Figure 3-b. The built e-nose.

In this study as distinct from similar studies, tea odour was not provided by occurring from a certain amount of the tea sample put into a box; tea odour was provided by transferring from above the fermentation band to the electronic nose. It has been considered that other odours could be mixed into the tea odour, so the tea odour was transferred from only 10 cm above the tea level, under 30 cm from the top of the side walls of the fermentation band. Thus, effects of other odours to the tea odour could be minimized.

Samples quantity is limited by 64, due to the cost of chemical analysis.

3. CLASSIFICATION PROCESS

Tf/tr chemical analysis was made from the sniffed tea samples. The tea was classified into 3 sub-groups according to tf/tr level;

- high quality tea,
- mid quality tea,
- low quality tea.

Every sniffing sequence took 60 seconds and 3000 samples were taken during one sequence. Fourteen outputs taken from 13 sensors were sampled. The TGS-2201 has 2 outputs. A matrix with size [64x14x3000] was filled with 64 samples. Then this matrix was down sized [64x14x1500] by eliminating the data sampled from the sensor cleaning and odour lock states to simplify the classification process.

According to the tf/tr values of 64 samples we had 19 samples with low quality, 21 samples with mid quality and 24 samples with high quality. Data were used 50% as training and 50% as test.

3.1. Feature Extraction Methods

Six different extracted features from the signal recorded by the e-nose are;

$$F1 = Sum(X) = \sum_{i=1}^{n}(x_i) \tag{2}$$

$$F2 = Mean(X) = \frac{1}{n}\sum_{i=1}^{n}(x_i) \tag{3}$$

$$F3 = Var(X) = \frac{1}{n}\sum_{i=1}^{n}(x_i - \mu)^2 \tag{4}$$

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$$F4 = Skewness(X) = \frac{\frac{1}{n}\sum_{i=1}^{n}(x_i - \mu)^3}{(\sqrt{\frac{1}{n}\sum_{i=1}^{n}(x_i - \mu)^2})^3} \tag{5}$$

$$F5 = Kurtosis(X) = \frac{E(x-\mu)^4}{\sigma^4} \tag{6}$$

In these equations x is the recorded tea signal, n is the length of x, μ is the mean of x, E is the expected value of the quantity and i represents the index of x, where i=1,2,3... n.

The singular value decomposition equation for an X (mxn) singular matrix A is:

$$F6 = A = U\Sigma V^T \tag{7}$$

Where U is an (m×m) orthogonal matrix, V is an (n×n) orthogonal matrix and Σ is an (m×n) diagonal matrix containing the singular values of A arranged in decreasing order of magnitude.

3.2. Linear Discriminant Analysis Classification Method

LDA classifies two classes based on the assumption that both classes are under normal distribution with equal covariance matrices. The separating hyper plane is obtained by finding the projection of the labelled training data that maximizes the distance between the two classes' means and minimizes the interclass variance. The main aim is to solve the problem:

$$y = w^T x + w_0 \tag{8}$$

where x is the feature vector. The vectors w and w_0 are determined by the maximization of the interclass means and minimization of interclass variance [22, 23].

3.3. Naïve Bayes Classification Method

Naïve Bayes classifier is a simple probabilistic algorithm based on applying Bayes' theorem with naïve independence assumptions. Consider a set of training trials where each trial is made up from m discrete-valued features and a class from a finite set C. The naïve Bayes classifier can probabilistically predict the class of an unknown trial using the available training trial set to calculate the most probable output. The most probable class C_{NB} of an unknown trial with the conjunction $A=a_1, a_2, ..., a_m$ is calculated by [23,24]:

$$C_{NB} = \arg\max_{c \in C} p(c \setminus A) \tag{9}$$

3.4. k-Nearest Neighbour Classification Method

The *k*-NN classifier is a common classification algorithm, which determines a testing sample's class by the majority class of the *k* closest training samples. Performance of a *k*-NN algorithm depends on the distance metric and the value of the closest training sample parameter, *k*. In our study, we used the Euclidean distance metric and we selected *k* parameter as 3, 5, 7 and 9 [22, 23].

4. EXPERIMENTAL RESULTS

Fourteen output values from 13 sensors are evaluated and 5 from all found as the most related with their conductivity values to tea fermentation odour (TGS-826, TGS-2104, TGS-2201-1. output, TGS-2602 and TGS-2620). We also observed that TGS-813, TGS-825, TGS-830, TGS-880, TGS-2201-2. Output, TGS-2610, TGS-2611 and TGS-5042 sensors had very little reaction to the same odour, where TGS-2180 had none.

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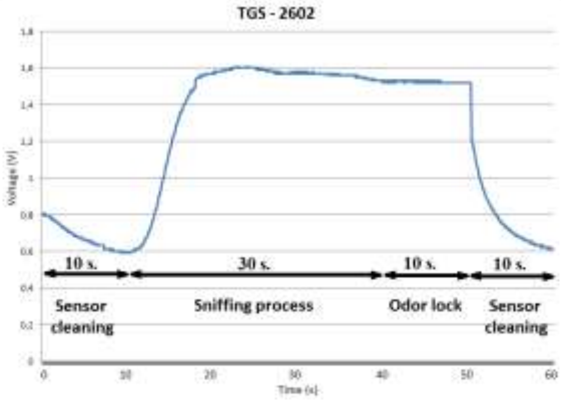

Figure 4-a. TGS-2602 sensor's voltage (V) vs. time for single sniffing cycle.

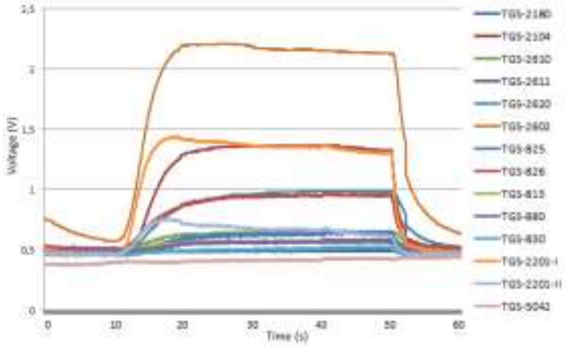

Figure 4-b. All sensors' voltages vs. time for single sniffing cycle.

Figure 4-a shows the voltage value vs. time for TGS-2602 sensor's single sniffing cycle and Figure 4-b shows the all sensors' voltage values vs. time. A radar graph of the sensor values for different tea odours can be seen in Figure 5-a and sensors voltage values' variance for single sniffing cycle can be seen in Figure 5-b.

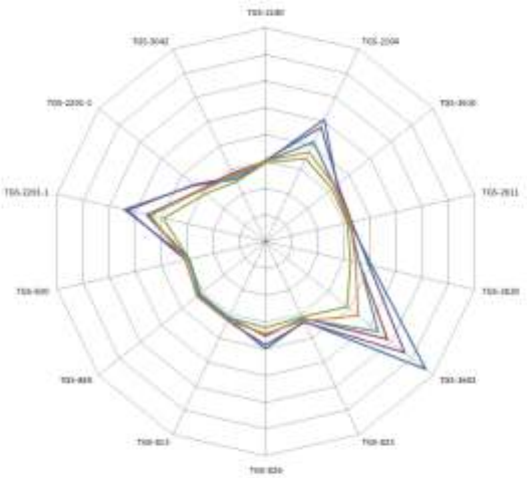

Figure 5-a. Radar graph of sensor values for different tea odours.

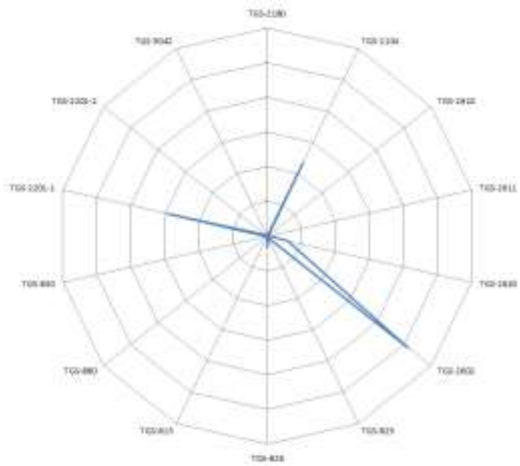

Figure 5-b. Sensors voltage values' variance for single sniffing cycle.

Figure 6 also shows TGS-2602 sensor's conductivity change vs. time for different moments of the fermentation process. As seen in Figure 6, TGS-2602 sensor has max-output around 1.8 V at early stages of fermentation while it has 1.1 V at late stages. Different output values for different qualities are essential for classification.

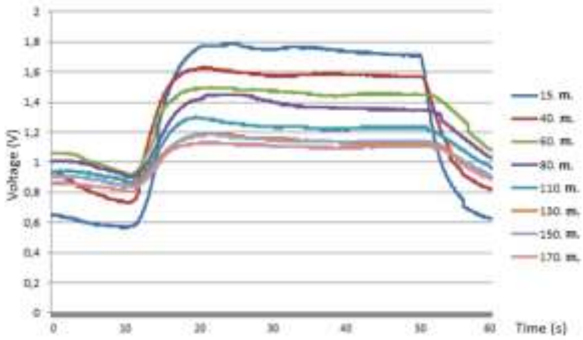

Figure 6. TGS-2602 output values during different moments of the fermentation process.

Table 2 shows the classification results with all algorithms we used. *k*-NN-3 has the highest accuracy for classifying the tea quality via an online e-nose system.

Table 2. Classification performances vs. algorithms.

k-NN-3	:	74.19 %
k-NN-5	:	64.52 %
k-NN-7	:	48.39 %
k-NN-9	:	45.16 %
LDA	:	41.94 %
Bayes	:	35.48 %

LDA algorithm has the second lowest performance because of its linear structure. Bayes has the lowest performance because the samples are nested, *k*-NN-3 gives the highest performance for this study with 74.19%.

5. DISCUSSION

While classification performances are 90% in other similar studies, the classification performance has been achieved at 74.19% in this study. The classification performance is lower than other studies on this point of view; however this practice was applied on a production band of a factory in real time. Factory officials stated that the classification performance was acceptable ranges. It is considered that the reasons of obtaining lower classification performance were because of not transferring odour from a certain amount of tea in a closed chamber to the e-nose and not using an air mass flow controller. So it might differ in odour input of the e-nose even though it is a small amount.

6. CONCLUSION

A 13 sensor e-nose has been built and 64 online different odour samples were taken from tea the fermentation line in a tea production facility in Rize which is a city of Turkey. A certain amount of tea was dried right after all sniffing processes and chemical analyses have been made (tf/tr) for their quality. Tea was classified into 3 groups as high, mid and low quality according to 64 tf/tr values, 50% of the datas were used as training and the other 50% as test. A data matrix with size of [64x14x3000] was filled with 64 different tea odours. According to sensor output values, only 5 of these 14 outputs were found relevant to the tea fermentation odour.

This study performed an online tea quality test experiment using an e-nose, achieving a 74.19% classification performance and successfully applied in the factory. An online quality test system exhibits that better quality tea production can be provided with some simple modifications on factory's automation.

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