Impact of Sensing Film's Production Method on Classification Accuracy by Electronic Nose

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Abstract: The development of gas sensing materials is relevant in the field of non-invasive biodevices. In this work, we used an electronic nose (E-nose) developed by our research group, which possess versatile and unique sensing materials. These are gels that can be spread over the substrate by Film Coating or Spin Coating. This study aims to evaluate the influence of the sensing film spreading method selected on the classification capabilities of the E-nose. The methodology followed consisted of performing an experiment where the E-nose was exposed to 13 different pure volatile organic compounds. The sensor array had two sensing films produced by Film Coating, and other two produced by Spin Coating. After data collection, a set of features was extracted from the original signal curves, and the best were selected by Recursive Feature Elimination. Then, the classification performance of Multinomial Logistic regression, Decision Tree, and Naïve Bayes was evaluated. The results showed that both spreading methods for sensing films production are adequate since the estimated error of classification was inferior to 4 % for all the classification tools applied.

1 INTRODUCTION

An electronic nose is a device able to detect and identify odours. The way it works is inspired in the mammalian's olfactory system (Sankaran et al., 2012), having two main parts: the perception and the recognition. The perception instrument has a delivery system (to carry the gas sample from the sample to the detector), a detection system with a sensor array (where the interaction with the gas sample occurs), and a transduction and data collection system (that converts the properties changing in the sensors into electrical signals) (Peris and Escuder-Gilabert, 2009). And the recognition part includes mathematical methods for features extraction and selection, as well as algorithms for pattern recognition (Yan et al., 2015).

The sensors might be based on Surface Acoustic Wave (SAW), Quartz Crystal Microbalance (QCM), Conducting Polymers (CP), Metal Oxide Semiconductors (MOS), optical sensors, among others (Gutiérrez and Horrillo, 2014). Each sensor reacts to the presence of the odour in a different way, depending on its own sensibility and specificity. The patterns of odours generated, also called "fingerprint", are electrical signals that result from the variance of the sensor properties, such as conductance, voltage and capacity. Furthermore, the signals acquired are analysed, the best features are scored, and a pattern recognition method is applied. For instance, Principal Component Analysis (PCA) has been employed to reduce the number of features extracted from the signals (Ghasemi-Varnamkhasti et al., 2015), (Ordukaya and Karlik, 2017). Additionally, machine learning algorithms are commonly used for odours recognition (Phaisangittisagul et al., 2010), (Zhang et al., 2008), and (Barbri et al., 2009) , and are also employed for concentration prediction (Xu et al., 2016).

Machine learning algorithms are becoming very important for medical data analysis. For instance, Decision Tree is a transparent and easily interpretable method, presenting a high potential for diagnostic model building. (Polaka et al., 2017).

We created a home-made E-nose based on optical sensors, which possess a new class of sensing ma-

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terials invented by our research group. These gels are very versatile and have unique stimuli-responsive properties, altering their optical configuration while interacting with volatile organic compounds (VOCs) (Hussain et al., 2017). This paper uses the E-nose system we developed, which was previously described in (Padua et al., 2018), for identification of 13 different pure VOCs. Two different spreading method techniques have been used to produce our sensing films: Film Coating and Spin Coating. In this study, we aim to test if sensing films produced by different spreading techniques have distinct VOCs discrimination capabilities.

Moreover, we were also interested in applying different machine learning algorithms for VOCs classification.

Support Vector Machine (SVM) for E-noses with MOS sensors array (Ghasemi-Varnamkhasti et al., 2015); and Neural network and the Large Margin Nearest Neighbours (LMNN) for E-noses using SAW sensors array (Hotel et al., 2018) produced the best classification results. However, when discrimination can be done with a simple method, the usage of more complex approaches is not needed (Ghasemi-Varnamkhasti et al., 2015).

(Ordukaya and Karlik, 2017) compared the performances of different machine learning algorithms for olive oil classification by an E-nose (*Cyranose 320®*): Naïve Bayes, k-Nearest Neighbours (k-NN), Linear Discriminant Analysis (LDA), Decision Tree, Artificial Neural Network (ANN), and SVM using two different approaches: using dimensional reduction and without data reduction. The best success rate was found with Naïve Bayes classifier after data reduction from 32 inputs to 8 inputs based on PCA.

The work of (Cho and Kurup, 2011) showed that decision tree models have excellent results for classification and provide easily interpretable tree structure for E-nose data. The decision tree approach was reported as a promising pattern recognition method with accuracy rates above 97 % using several features extracted from signals acquired by an E-nose based on MOS sensors. The training of the decision tree was also faster compared to Multilayer Perceptron (MLP) and fuzzy ARTMAP classifiers.

(Siavash et al., 2018) developed a research where used E-noses based on MOS sensors (*FOX 400, Alpha M.O.S, France*) and Field-Asymmetric Ion Mobility Spectrometry (*FAIMS, Owlstone Lonestar*) to distinguish healthy from diabetic patients. High prediction accuracy was achieved by combining PCA with a sparse logistic regression and a Gaussian process classifier. Another study used Logistic regression to classify data collected by an E-nose to identify correctly biofilm-producing versus non biofilm-producing bacteria species with accuracy ranging from 72.2 % to 100 %, depending on the organism and methodology. When the dataset is not complex, simple methods might be better than complicated techniques to discriminate between different classes (Ghasemi-Varnamkhasti et al., 2015).

We decided to test three simple machine learning algorithms, since other authors have reported the capability of other E-noses to perform classification of samples in a fast and reliable way using those methods (Ordukaya and Karlik, 2017) (Ghasemi-Varnamkhasti et al., 2015). In case we were not able to achieve good performance, we intended to apply more advanced computational techniques to enhance classification accuracy.

Taking into account the results obtained in the previous studies, our study was focused on the performance of Logistic Regression, Decision Tree and Naïve Bayes as data classifier methods. Finally, the class discrimination capabilities of these classifiers were compared.

2 MATERIALS AND METHODS

2.1 Data Collection

A schematic of the E-nose system used is represented in Figure 1. The device is composed of three main parts: the delivery system, the detection chamber and the transduction system.

The delivery system has two pumps working alternately: the exposure pump, which creates a gradient of pressure that transfers the headspace from the sample chamber to the detection chamber; and the recovery pump, that is responsible for purging the detection chamber (in the recovery periods).

The detection chamber is where the interaction of the VOCs with the array of sensors takes place. Here, the optical sensors that compose the array change their optical properties while interacting with VOCs, and these changes are detected by Light Dependent Resistors (LDRs) and converted into electrical signals.

Signals' collection is ensured by the transduction system. It is composed of a microcontroller (Arduino Due), that converts the analogue signals into digital signals, and an embedded system (Raspberry Pi 2 Model B), for signals collection. More details about the device have been earlier reported (Padua et al., 2018).

Inside the detection chamber, six optical sensors based on tunable sensing films were used in the sensor array - see Figure 2 a).

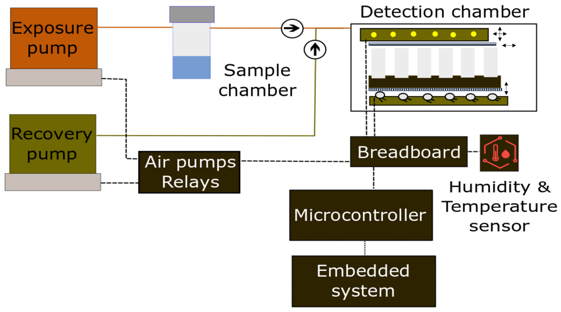


Figure 1: Schematic representation of E-nose V2.

Each sensor is composed of:

- a source of unpolarised light - a Light Emitting Diode (LED);
- a sensing film - based on tunable sensing gels described in (Hussain et al., 2017) - sandwished between two crossed polarising filters;
- a Light Dependent Resistor (LDR).

As shown in Figure 2 b), a sensing film consists of a thin layer of sensing gel spread on a glass slide with a black mask on the back (that has a 5 mm diameter hole, which delimiters the area of detection). The sensing gels' composition is based on a biopolymer matrix with droplets of liquid crystal (LC), self-assembled in the presence of ionic liquid (IL) (Hussain et al., 2017).

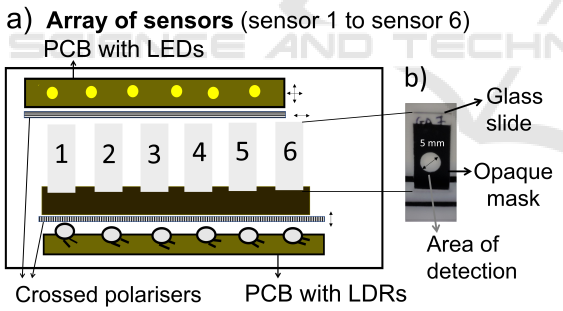


Figure 2: a) Schematic of sensor array. ; b) Sensing film.

In the present study, sensors nr 4 and 5 have sensing films produced by a standard (STD) recipe described in Table 1, and sensors 1 and 3 are also produced by the standard recipe, plus adding 1.5 μL of a cross-linking agent to the standard recipe, named glutaraldehyde (GA), for 5 min at 500 RPM magnetic stirring. Sensors 2 and 6 are controls: sensing film 2 does not have IL and sensing film 6 does not contain LC.

After the gel's production, the sensing films might be spread on the glass slides by two different film applicator equipment: Film Coater or Spin Coater. The Film Coater used was the automatic film applicator with perforated heated vacuum bed from *TQC Sheen*) - Figure 3 a), and the Spin Coater used was the SPIN150i Tabletop from *POLOS* - Figure 3 b).

Table 1: Standard recipe for sensing films production.

Component	Time (min)	Magnetic Stirring (RPM)
BMIM DCA (IL)	10	300
5 CB (LC)	10	300
BSG	10-15	500
MilliQ water	5	500

IL = Ionic Liquid; LC = Liquid Crystal ; BSG = Bovine Skin Gelatin.

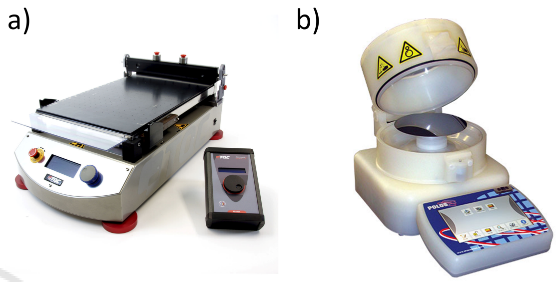


Figure 3: a) Film Coater ; b) Spin Coater.

The thickness of the sensing films produced by Film Coating is 30 μm, and the drop quantity of gel used was 15 μL. Finally, the optical sensing films with light polarisation properties were obtained.

For this experiment, in the sensor array, sensing films 1 and 4 were spread by Film Coating (FC), whereas sensing films 3 and 5 were spread by Spin Coating (SC). A brief description of each sensor is provided in Table 2.

The experiment performed consisted in exposing the same array of sensors (Table 2), placed inside the detection chamber, to a set of 13 VOCs, sequentially, in the following order: acetone, isopropanol, ethanol, methanol, hexane, heptane, toluene, xylene, benzene, chloroform, dichloromethane, diethyl ether, and ethyl acetate. The experiment conditions are described in the list below:

- **VOC quantity:** 20 mL
- **Sample temperature:** 37 °C
- **Time exposure:** 5 s
- **Time recovery:** 15 s
- **Duration:** 20 min
- **Sampling rate:** 5 Hz

The data was collected by the E-nose data transduction and acquisition system. An example of a set of 6 signals (one per sensor) acquired for exposure to acetone is shown in Figure 4.

Table 2: Sensor array of the E-nose.

Sensor	Sensing film description	Spreading technique
1	STD gel + GA	FC
2	Control (without IL)	FC
3	STD gel + GA	SC
4	STD gel	FC
5	STD gel	SC
6	Control (without LC)	SC

STD = Standard ; GA = Glutaraldehyde ; FC = Film Coating ; SC = Spin Coating ; LC = Liquid Crystal ; IL = Ionic Liquid.

2.2 Data Analysis

After data collection, the features related to each individual sensor were extracted. Table 3 describes the 8 features extracted per sensor, and Table 4 explains the physicochemical meaning of each feature. Since the sensor array is composed of 6 sensors, the number of original features that can be used is given by $6x8 = 48$.

Then, auto-scaling was exploited for data preprocessing, using a standardisation technique, according to Eq. 1:

$$Z_j = \frac{(x_j - \bar{x_l})}{\bar{s_l}} \tag{1}$$

where Z_j is the value of x_j after auto-scaling. x_j is defined as the variable before scaling. $\bar{x_l}$ is the variable mean and $\bar{s_l}$ is the standard deviation of the variable. The final value Z_j varies around the mean zero with standard deviation one.

2.2.1 Features Selection

We were interested in comparing the optical responses given by sensors where sensing films that were spread by Film Coating were applied, versus from sensors with sensing films that were spread by Spin Coating.

Therefore, the original set of 48 features was divided into two sub-groups (see Figure 5). One group is composed by 16 features of sensing films produced by Film Coating (sensors 1 and 4 - red signals in Figure 4), and the other is composed by 16 features of sensing films produced by Spin Coating (sensors 3 and 5 - blue signals in Figure 4).

For each group of features, we were interested in knowing the best number of features to select, and what features were more interesting for VOCs classification.

Initially, each sub-group has 16 features. To know the more relevant features for differentiating the VOCs, Recursive Feature Elimination (RFE) was performed. The estimator 'svc' was used to assign weights to features. This estimator was trained on the initial set of features and the importance of each feature was obtained. Then, the RFE method removes the weakest features and selects them by recursively considering smaller and smaller sets of features. The procedure is recursively repeated on the pruned set until a specific number of features to select is reached. For each sub-group of features, RFE was used to select the best number of features, and the top best were selected by cross-validation (2 folds).

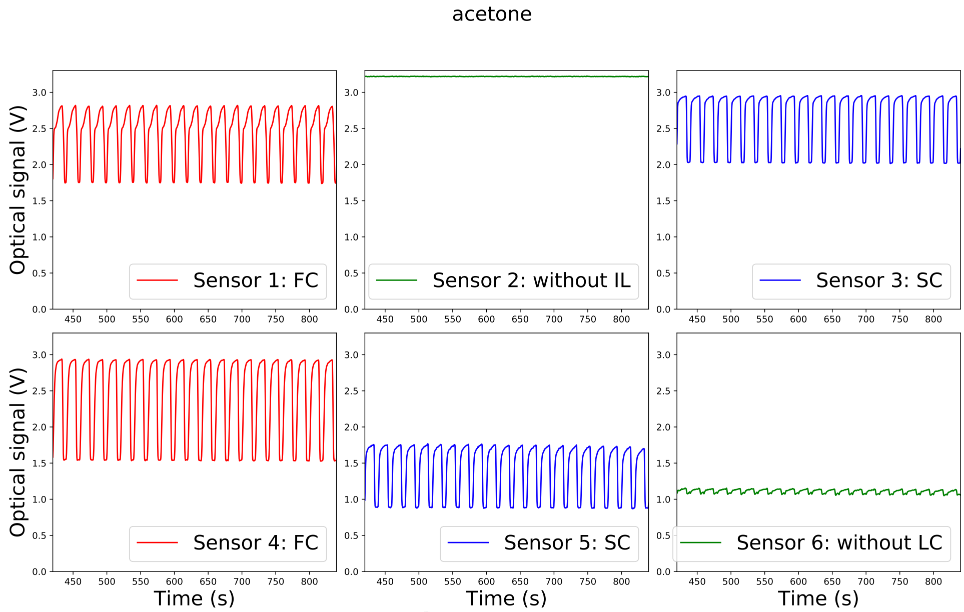
2.2.2 Classification Models

We studied which classification model works best for sensors with sensing films produced by Spin Coating and Film Coating. To asses the quality of various prediction methods, we trained models with three different classification techniques: Multinominal Logistic regression, Decision Tree, and Naïve Bayes.

Figure 6 shows the procedure used for each classifier. Each group of best features related to Film Coating or Spin Coating was randomly divided in train data (70 %) and test data (30 %).

Cross-validation (10 folds) was applied on the train data. This means that the train set was randomly divided in 10 subsets, using 9 for training the model and the remaining to validate it. Firstly, the model was built on the train set, then the training error was calculated; the validation set was tested separately and the validation error was also obtained. This procedure was repeated 9 more times, each time using a different subset for validation. The average over classes of cross-validation for the different classification techniques was reported. The parameters of the models associated to a lower validation error were selected for classifiers optimisation.

The parameters of optimisation were then applied on the classifier for prediction on the Test data. Finally, for each classifier, the estimated error was calculated, and the McNemar's test was used to compare the classifiers.



FC: sensor with sensing film spread by Film Coating; SC: sensor with sensing film spread by Spin Coating; IL: Ionic Liquid; LC: Liquid Crystal.

Figure 4: Examples of cycles from signals collected when the sensor array is cyclically exposed to acetone for 5 min, during the 20 min experiment.

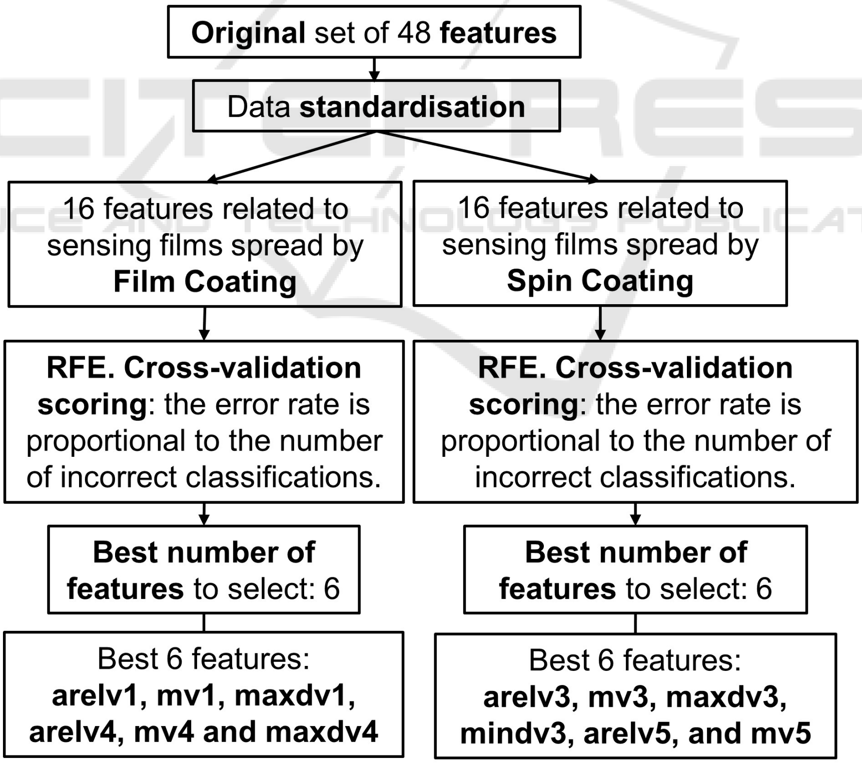


Figure 5: Schematic of features selection algorithm.

2.2.3 Naïve Bayes

Naïve Bayes methods are a set of probabilistic algorithms based on applying Bayes theorem with strong independence assumption between every pair of features.

For this work, we assumed the likelihood of the features to be Gaussian. We applied the *GaussianNB*

Table 3: Features extracted from the signals.

Feature name	Description
arelv (+ nr sensor)	signal's relative amplitude
amax (+ nr sensor)	x coordinate of signal's maximum
max (+ nr sensor)	y coordinate of signal's maximum
amaxdv (+ nr sensor)	x coordinate of maximum of signal's first derivative
maxdv (+ nr sensor)	y coordinate of maximum of signal's first derivative
amindv (+ nr sensor)	x coordinate of minimum of signal's first derivative
mindv (+ nr sensor)	y coordinate of minimum of signal's first derivative
onset (+ nr sensor)	time at which the signal raises above the noise after the recovery time have started

Table 4: Features meaning.

Feature name	Meaning
arelv (+ nr sensor)	Value influenced by concentration of VOC and affinity towards sensing gel
amax (+ nr sensor)	Time needed for maximum detection of VOC
max (+ nr sensor)	Level of maximum VOC detection
amaxdv (+ nr sensor)	Time when the rate of interaction VOC-sensing film is higher
maxdv (+ nr sensor)	Maximum rate of interaction VOC-sensing film
amindv (+ nr sensor)	Time when the rate of unlink VOC-sensing film is higher
mindv (+ nr sensor)	Maximum rate of unlink VOC-sensing film
onset (+ nr sensor)	Time needed for the sensor to start the response

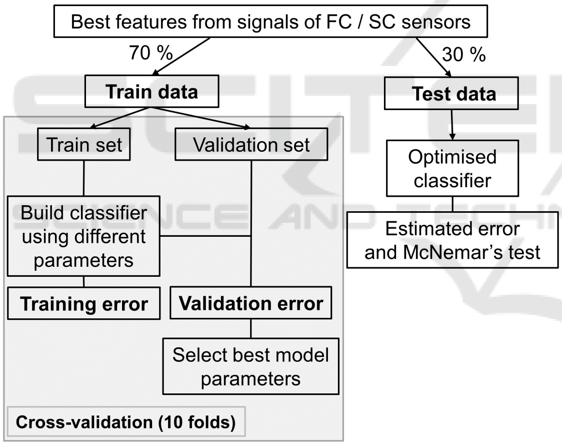


Figure 6: Schematic of classifiers' algorithm.

function from the *scikit-learn* library to implement the Gaussian Naïve Bayes algorithm for classification.

2.2.4 Decision Tree

A Decision Tree is a flowchart, where each branch represents the outcome of a decision, and each terminal node holds a class label. It is a method simple to understand, interpret and visualise data.

The Decision Tree applied was the *DecisionTreeClassifier* from *scikit-learn* library, and its best maximum depth was studied.

2.2.5 Multinomial Logistic Regression

Multinomial logistic regression generalises logistic regression to multiclass problems, i.e. with more than two possible discrete outcomes.

We used the Logistic regression classifier from the *scikit-learn* library. The one-vs-all methodology was applied. Our dataset is composed by several classes, therefore we need to decompose our training set into 13 different binary classification problems, where one of the classes/labels corresponds to 1, and the remaining labels correspond to 0. Each logistic regression classifier is defined by:

$$h_{\Theta}^{(i)}(x) = P(y = i \mid x; \Theta), i = (1, 2, ...13) \quad (2)$$

We train the logistic classifier for each class i to predict the probability of an input y that $y = i$. When we want to predict a new input x, we pick the class that maximises the probability of x belonging to a certain class:

$$max_i\, h_{\Theta}^{(i)}(x) \quad (3)$$

3 RESULTS

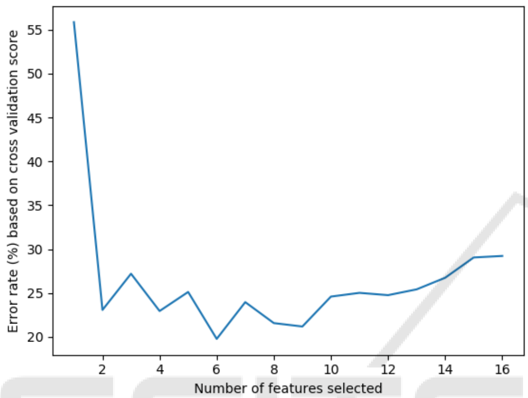
3.1 Features Selection

According to RFE - see Figure 7a - the lower error rate of the classifier with 2-fold cross-validation occurs for

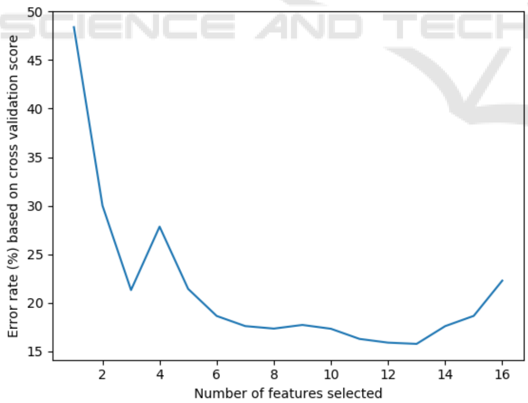
6 features. Consequently, the best number of features for VOCs classification using sensors with sensing films produced by Film Coating is 6.

Figure 7b shows that the best number of features for VOCs classification using sensors with sensing films produced by Spin Coating is also 6. A higher number of features decreases the error rate. However, the error decreases less than 5 % using 13 features. Since a lower number of features improves the computation performance, we decided to use also 6 features for this group of sensors.

Therefore, in the next steps, only the best 6 features per group were selected.



(a) Features extracted from signals obtained from sensors with sensing films produced by Film Coating.



(b) Features extracted from signals obtained from sensors with sensing films produced by Spin Coating.

Figure 7: Correlation between cross-validation scores of RFE vs number of features extracted from signals.

The ranking obtained by RFE indicated that the 6 features with a higher score were: arelv1, mv1, maxdv1, arelv4, mv4 and maxdv4 for Film Coating; and arelv3, mv3, maxdv3, mindv3, arelv5, and mv5 for Spin Coating. Therefore, these features were the only used for training and testing in the classification models performed.

3.2 Classification

We studied the best parameter C for Logistic regression, and the best maximum depth of Decision Tree, using features extracted from the signals given by sensors where sensing films produced by Film Coating were used, and by sensors where sensing films produced by Spin Coating were used. Then, we assessed the accuracy of the three classification models: Logistic regression, Decision Tree and Naïve Bayes.

3.2.1 Classification using Sensors with Sensing Films Produced by Film Coating

The value of parameter C for Logistic Regression was optimised - Figure 8a , as well as the best value for depth in Decision Tree - Figure 8b.

Using sensing films produced by Film Coating in the sensors, the best value of parameter C for Logistic Regression is 65536 - Figure 8a. And the best depth for Decision Tree is 15 - Figure 8b. Hence, these values were selected to be used in the classifiers' models.

The estimated errors calculated on the Test set were 2.75 % for Multinominal Logistic Regression, 3.15 % for Decision Tree, and 3.15 % for Naïve Bayes.

We also compared the difference in accuracy between the classifiers using the McNemar's test, which indicates a significant difference between two classifiers (with 95 % confidence) if the value of the test is ≥ 3.84. The results for Film Coating were the following:

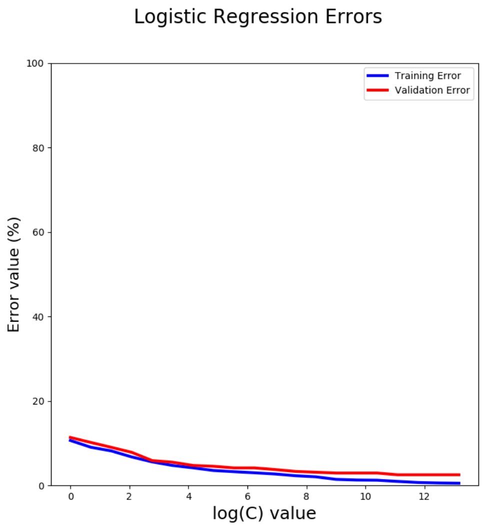
- Multinominal Logistic Regression vs Decision Tree: 0.0
- Multinominal Logistic Regression vs Naïve Bayes: 0.0
- Decision Tree vs Naïve Bayes: 0.1

The results above indicate that there is no significant difference between the accuracy of the three classifiers used.

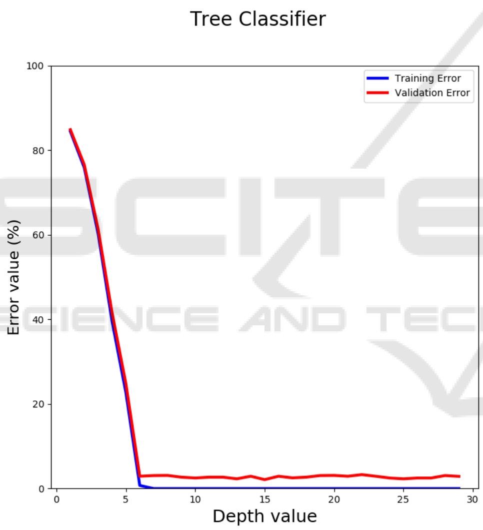
3.2.2 Classification using Sensors with Sensing Films Produced by Spin Coating

Using sensing films produced by Spin Coating in the sensors, the best value of parameter C for Logistic Regression is 32768 - Figure 9a. And the best depth for Decision Tree is 8 - Figure 9b. Hence, these values were selected to be used in the classifiers' models.

The estimated errors calculated on the Test set were 2.76 % for Multinominal Logistic Regression, 3.15 % for Decision Tree, and 3.94 % for Naïve Bayes.

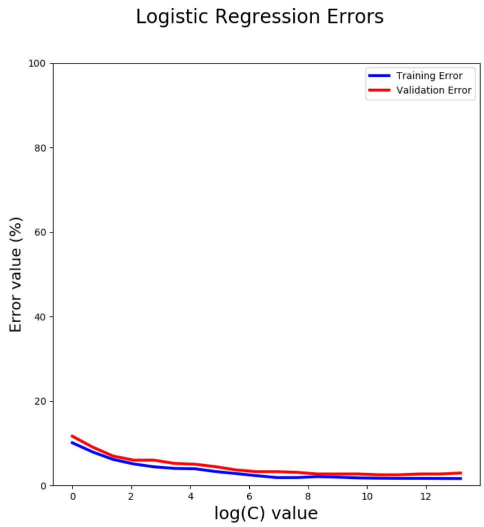


(a) Optimisation of C parameter for Logistic Regression.

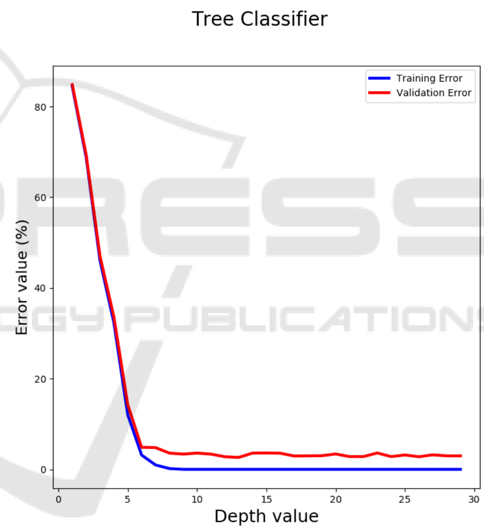


(b) Optimisation of depth parameter for Decision Tree.

Figure 8: Optimisation of algorithms’ parameters using sensing films produced by Film Coating.



(a) Optimisation of C parameter for Logistic Regression.



(b) Optimisation of depth parameter for Decision Tree.

Figure 9: Optimisation of algorithms’ parameters using sensing films produced by Spin Coating.

The McNemar’s test was also performed. The results for Spin Coating were the following:

- Multinominal Logistic Regression vs Decision Tree: 0.00
- Multinominal Logistic Regression vs Naïve Bayes: 0.36
- Decision Tree vs Naïve Bayes: 0.08

The results above indicate that there is no significant difference among the accuracy results of Multinominal Logistic Regression, Decision Tree and Naïve Bayes.

4 CONCLUSIONS

In this study, the RFE results indicate that the most effective features for classifications are: relative amplitude, maximum of the signal, and maximum and minimum of signal’s first derivative.

For distinction of 13 different VOCs, the three simple classification methods studied were effective, with estimated error inferior to 4 % for all of them.

Comparing the results obtained for sensors with sensing films produced by Film Coating vs sensing films produced by Spin Coating, the values did not vary significantly for any of the classifiers. Therefore, we conclude that both spreading method techniques are very good for sensing films production, and none of them revealed to be better than the others for VOCs classification.

The E-nose system and the machine learning algorithms applied in the present study demonstrated capability to distinguish the different VOCs in a quick, simple and accurate way, using both sensing film production types.

Future studies can be performed in order to explore the application of the E-nose in many different sectors, such as food and beverage evaluation, environmental safety or medical research. Moreover, other Machine Learning algorithms can be explored and optimised.

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