RECURSIVE LEAST SQUARES ALGORITHM DEDICATED TO EARLY RECOGNITION OF EXPLOSIVE COMPOUNDS THANKS TO MULTI-TECHNOLOGY SENSORS

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ABSTRACT

In this paper, a novel gas identification approach based on the Recursive Least Squares (RLS) algorithm is proposed. We detail some adaptations of RLS to be applied to a sensor matrix of several technologies in optimal conditions. The low complexity of the algorithm and its ability to process online samples from multi-sensor make the real-time identification of volatile compounds possible. The effectiveness of this approach to early detect and recognize explosive compounds in the air has been successfully demonstrated on an experimentally obtained dataset.

Index Terms— Electronic nose, Pattern recognition, Multidimensional analysis, Recursive Least Squares

1. INTRODUCTION

In recent years, there has been a growing interest in the development of gas sensors for the detection, identification or quantification of volatile compounds. This surge has been mainly driven by a variety of real-life applications such as air quality control for environmental protection or gas leakage monitoring for the prevention of industrial accidents. A multitude of sensor technologies has been developed for these applications including semiconductor, piezoelectric, optical, catalytic and electrochemical gas sensors [1]. However, all sensors have the same functioning principle which is based on a physicochemical interaction between the gaseous compound and a chemically sensitive material coated on the surface of the sensor and interfaced to a transducer [2]. In this way, the gas molecules, which represent the inputs, are absorbed onto the surface of the sensitive material involving some physical changes (temperature, optical, conductivity, mass...) which are sensed by the transducer (via changes in current, voltage, resistance, oscillation frequency, light fluorescence... depending on the technology) and converted to an output signal $x(t)$ at a given time t (Fig : 1). Thus, the output pattern depends on the used technology, the sensitive material and of course the analyte.

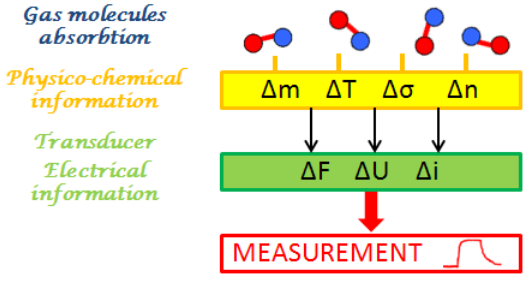


Fig. 1. Illustration of gas sensor principle.

In practice, a gas sensor array (different technologies and/or sensitive materials) is usually used instead of a single gas sensor to improve the selectivity of the whole system and the ability to classify several compounds. Thus, the output of the system $\mathbf{x(t)}$ is a multivariate signal. A signal processing step is then necessary to take a decision about the presence, the nature and the concentration of a targeted compound in the air. Such an intelligent system (gas sensors + signal processing) is called electronic nose (e-nose).

The signal processing techniques aim at finding a relationship between the sensors outputs and a set of reference signatures (or models) which characterize the expected response of each odor to be identified. This problem remains challenging for real-life applications of electronic noses due to the large intra-class variability compared to a thin inter-class separation and the small amount of data available. This is typically the case in the gas sensor area where it is very costly and time-consuming to obtain a large and representative set of examples in order to build reliable reference signatures.

Among numerous approaches proposed in the literature to process signals from e-noses, the most common procedures, based on multivariate statistics and neural networks [3, 4], exploit features from the steady state of the sensors response. As the number of training examples is often small relative to the number of features, it is noted that a dimensionality reduction technique (PCA [5], LDA [6]...) is applied before the classification step. The main drawback of these methods is the slowness of their decision because the process has to wait for the steady state to be reached. These systems are therefore not appropriate for the early detection of dangerous gases, such as explosives. Furthermore, as the transient stage is ignored, these approaches do not take advantage of reponse kinetics to discriminate gas fingerprints.

Alternate solutions have been proposed to overcome this issue. On the one hand, authors in [7] extract features from both dynamic and steady state data. Thus, performance of the classification system (based on neural networks) are improved but the authors do not care about having an early identification. On the other hand, the approach proposed in [8] is based on a time-delay neural network which performs dynamic features extraction thanks to a temporal sliding window. Data are therefore analyzed every second from 0 to 4min but decision about the nature of the compound is only taken at the end of the processing. Consequently, this approach is not suitable for real-time applications which require an early identification.

In this paper, a dynamic parameter estimation technique based on the Recursive Least Squares (RLS) algorithm is proposed as an alternative to common features extraction techniques in order to assure an early identification of gas compounds in the air. Algorithm's implementation and performance will be discussed and illustrated in the context of a multi-technology e-nose used for real-time detection, recognition and quantification of explosive compounds.

2. CONTEXT

With the increased use of explosives in terrorist attacks, law enforcement agencies are faced with the problem of detecting hidden bombs

in luggage, cars and aircrafts. To cope with the terrorist threat, the development of efficient e-noses has become an urgent worldwide necessity. Such a device should be dedicated to the identification of a large set of explosive compounds. Moreover, the constraint about response time is critical because the presence of a threat in the air should be detected and then identified in a few seconds.

In this paper, we propose a multisensor e-nose combining Fluorescence (OPTO) [9], Quartz Crystal Microbalance (QCM) [10] and Surface Acoustic Waves (SAW) [11]. These technologies have been chosen because they have different properties in terms of selectivity, sensitivty and response time. Absorption of compounds on the surface of the optical sensor induces a variation of light intensity whereas both piezoelectric sensors detect frequency variations. In order to improve the system selectivity and robustness, two different active layers are used for both piezoelectric sensors. In this way, the output of the sensor array is a five-dimensional signal (Fig : 2).

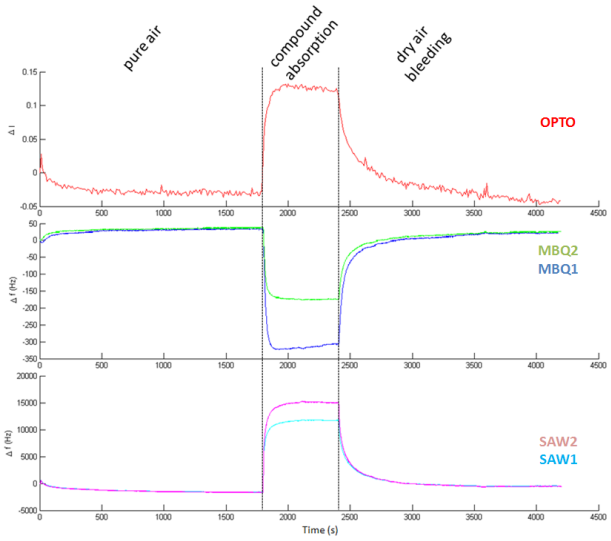


Fig. 2. Signal acquisition example obtained by our proposed multisensor e-nose.

Within the framework of explosives identification, our objective consists in recognizing two targets which are TNT and EGDN. The selectivity of the complete system will be estimated thanks to interferents such as ethanol (EtOH), dichloromethane (DCM) and methyl ethyl ketone (MEK).

3. REAL-TIME MULTI-SENSOR IDENTIFICATION

In order to process the multidimensional signal, we propose a method based on an hypothesis test approach built upon RLS algorithm combined with a decision step. This approach is based on an early fusion of the sensors signals and thus allows a positive synergy between the three technologies (response time, sensitivity and selectivity). This new approach is called Real-Time Multi-Sensor Identification (RT-MSI).

3.1. Set of equations

The proposed approach is based on the observation that the absorption of a gas compound on a chemical sensor can be modeled as a first order response (i.e. exponential function). Nevertheless, a linear drift estimation specific to each sensor has to be added to refine this simple model. Finally, the sensor response can be formulated with sufficient accuracy by Langmuir model [12] :

$$f_{\delta,\tau}(t,\theta) = Q\delta\left(1 - e^{-\frac{t}{\tau}}\right) + \alpha t + \beta, \tag{1}$$

where τ is the characteristic time of the first order response, and δ, the sensor sensitivity. These two parameters are characteristic of each compound response because they depend on the absorption affinity between the unknown gas and the sensor. The other parameters define the vector $\theta = (Q, \alpha, \beta)^T$. Q denotes the unknown compound concentration, α the slope of the sensor linear drift and β the sensor offset. This vector depends on the experimental conditions. In practice, for each sensor and each target (or hypothesis) c, $(\tau^{(c)}, \delta^{(c)})$ are set using theoretical basis and/or training examples in order to build models of the expected responses. Next, RLS algorithm estimates in real time θ in order to adjust the model of each hypothesis to best fit the real signal.

3.2. RLS algorithm

We now remind RLS principle [13] and we will detail specific modifications applied in order to adapt this algorithm to our issue. In the following, subscript c (which refers to the tested hypothesis) is omitted to simplify equations writing.

3.2.1. One dimensional signal case

Least Squares application corresponds to the search of the parameters vector θ minimizing the mean square error between the signal and the model which is in linear case :

$$\begin{aligned} Z &= H\theta + \epsilon \\ &= \begin{pmatrix} & \vdots & \\ \delta(1-e^{-\frac{t}{\tau}}) & t & 1 \\ & \vdots & \end{pmatrix} \begin{pmatrix} Q \\ \alpha \\ \beta \end{pmatrix} + \epsilon \end{aligned} \tag{2}$$

where Z denotes the signal vector, H the model matrix, θ the vector of parameters and ϵ the estimation error. Parameters which minimize mean square error between signal and model correspond to the pseudo-inverse solution $\hat{\theta} = (H^T H)^{-1} H^T Z$.

For real-time applications, RLS approach consists in computing the least squares estimate of θ with all measurements available at time $k+1$, but without recomputing the pseudo-inverse solution (which would be very expensive), but rather by updating the estimate available at k. Let's reformulate Equation (2) for explanations :

$$\begin{aligned} \begin{bmatrix} Z_k \\ z_{k+1} \end{bmatrix} &= \begin{bmatrix} H_k \\ h_{k+1} \end{bmatrix} \theta + \begin{bmatrix} \epsilon_k \\ \varepsilon_{k+1} \end{bmatrix} \\ \Leftrightarrow \quad Z_{k+1} &= H_{k+1} \theta + \epsilon_{k+1} \end{aligned} \tag{3}$$

RLS solution consists in updating θ with each new sample in a recursive manner (see [13] for further details) :

$$\hat{\theta}_{k+1} = \hat{\theta}_k + P_{k+1} h_{k+1}^T \left(z_{k+1} - h_{k+1}\hat{\theta}_k\right) \tag{4}$$

where $\hat{\theta}_{k+1}$ (respectively $\hat{\theta}_k$) is the estimation of θ at time $k+1$ (respectively k). Practical initialization is $\theta_0 = 0$ and $P_0 = Id$. Using Shermann-Morrison-Woodburry approach, P_k is updated in such a way :

$$P_{k+1} = P_k - \frac{P_k h_{k+1}^T h_{k+1} P_k}{1 + {h_{k+1}}^T P_k h_{k+1}} \tag{5}$$

The very low complexity of this algorithm allows a real-time use. In the next part, RLS algorithm is presented in the framework of multi-technology.

3.2.2. Multisensor adaptation

As introduced in [14], the most specific aspect of our application is the formulation of the regression for a multisensor system. The objective is to exploit the whole set of responses coming from the three technologies presented in section 2. On the one hand, it is important to formulate the model in order to take into account the common compound concentration Q. On the other hand, the slope α and the offset β take different values for each sensor, since the part of the model is precisely dependent on sensors. Considering equation (2) and merging all parameters in the same vector, monochannel models become in the case of a two channels acquisition :

$$Z^1 = H^1\theta + \epsilon^1 \quad (6)$$
$$= \begin{pmatrix} & \vdots & & & & \\ \delta_1(1-e^{-\frac{t}{\tau_1}}) & t & 0 & 1 & 0 & \\ & \vdots & & & & \end{pmatrix} \begin{pmatrix} Q \\ \alpha_1 \\ \alpha_2 \\ \beta_1 \\ \beta_2 \end{pmatrix} + \epsilon^1$$

$$Z^2 = H^2\theta + \epsilon^2 \quad (7)$$
$$= \begin{pmatrix} & \vdots & & & & \\ \delta_2(1-e^{-\frac{t}{\tau_2}}) & 0 & t & 0 & 1 & \\ & \vdots & & & & \end{pmatrix} \begin{pmatrix} Q \\ \alpha_1 \\ \alpha_2 \\ \beta_1 \\ \beta_2 \end{pmatrix} + \epsilon^2$$

Equations (6) and (7) allow formulating signals of both sensors acquiring simultaneously the same gas. Vector θ corresponds to the resolution of the equation implying sample concatenation with $Z = [Z^1 Z^2]^T$ and models concatenation with $H = [H^1 H^2]^T$. For the n^{th} sample acquired at time k and according to its original sensor i, the $(n)^{th}$ line of H, $h_n = (\delta_i(1 - exp(-t_k/\tau_i)) \dots 0 \dots t_k \dots 0 \dots 1 \dots)$, is built. It is noted that this formultation is suitable to accumulate samples from sensors with different sampling frequencies and we are not restricted by the number of channels (sensors).

Through this example, the ability to conceive a global model is illustrated. It formulates a correlated behavior between sensors (the exponential evolution linked to the first column of the model matrix) and in the same time a specific behavior for each one (the slope and the offset formulated through other columns of the matrix). From an identification point of view, this global model should be suitable to discriminate compounds with different kinetics and/or ampltitude ratio from the multi-sensor.

3.3. Decision

As explained in section 3.2, RLS algorithm provides, for each target c, a current error $e_k^{(c)} = \sqrt{\frac{1}{k}\sum_{i=1}^{k}(\varepsilon_i^{(c)})^2}$ which accounts for the gap between the model and the real data at time k. The smaller the gap is, the more probable is the presence of the associated compound in the air. For a better understanding, the current error is changed into probability (Eq.8) and a decision is taken as soon as a compound has its probability becoming greater than 65%.

$$P_k^{(c)} = \psi e^{-\left(\frac{e_k^{(c)}}{\sigma}\right)^2} \quad (8)$$

where ψ and σ are empirically chosen. In practice, to help the system to take the good decision, *a priori* knowledge is introduced and presented in the following.

3.3.1. Regularization

At the beginning of the absorption, the sensor drift and the exponential evolution cannot be discriminated correctly to enable a good estimation of parameters Q, α and β. So, in order to avoid confusion, we hypothesize that linear drift is low and a regularization has been formulated :

$$\hat{\theta} = \arg\min_{\theta} \left(\|H\theta - Z\|^2 + \|\Gamma\theta\|^2 \right) \quad (9)$$

where Γ is a diagonal matrix whose values are used to set each parameter inertial. The solution proposed by Tikhonov [13] is $\hat{\theta} = (H^T H + \Gamma^T\Gamma)^{-1} H^T Z$. In the case of RLS algorithm, solution of (9) implies $P_0 = (\Gamma^T\Gamma)^{-1}$.

3.3.2. Constraints

In practice, it does not make sense to have a negative concentration of compounds. A positivity constraint on this parameter is thus included in the analysis process. It takes place after the mean square error computation and not during convergence. It inhibits assumptions which provide a negative concentration through regression. In the same way, assumptions with a sensors drift whose the slope and/or the offset is big are ignored because the drift is expected to be slight.

4. EXPERIMENTS

In this section, we present experimentations which have been lead on real data with RT-MSI approach. The first part describes acquisition protocol whereas performance are discussed in the second part.

4.1. Protocol

All targets and interferents were acquired in lab condition (*i.e.* temperature, humidity and pressure were under control). Approximatively 1g of each compound was placed in a vapour generation cell and acquisitions were done for different concentrations ($h100 > h50 > h10$) by varying the absorption distance inside the cell (Fig : 3).

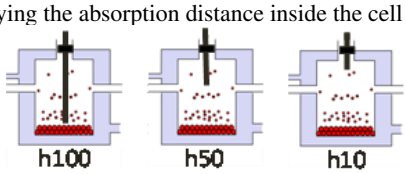
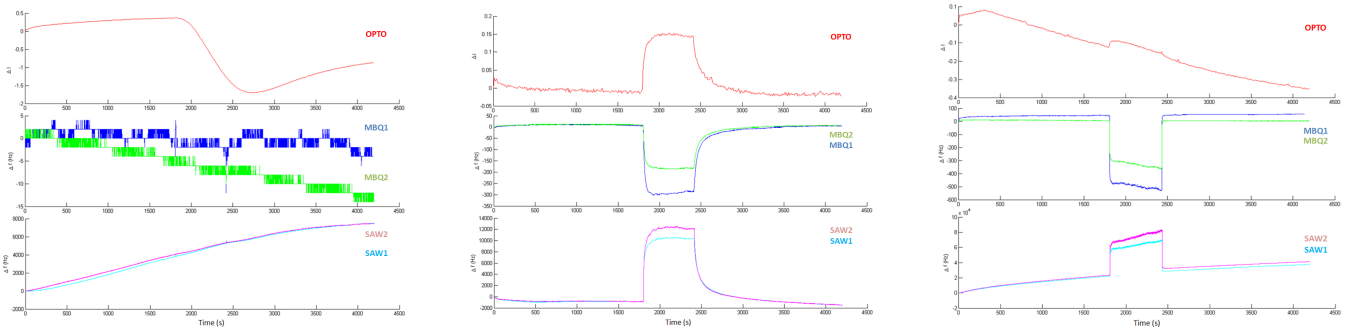


Fig. 3. Illustration of vapour absorption for different concentrations (h refers to the absorption distance inside the cell).

For experimentations, signals from sensors are sampled at different frequencies (10Hz for SAW, 1Hz for MBQ and 0.1Hz for OPTO), and the detection step is simply a threshold based approach on signals variance (calculated on a temporal sliding window).

4.2. Performance

Performance obtained by RT-MSI algorithm on real data are reported in Table 1. These satisfying results prove that the proposed e-nose is well adapted to identify explosive compounds in the air. Indeed the identification rate is very good (94%) and the recognition is always done less than 60s after the introduction of the compound in the air. The worst case appears for TNT (47s). This compound has the slowest kinetics as it is less volatile than the others (see TNT response vs. other responses on Fig : 4). Furthermore, RT-MSI algorithm is robust to variations of concentration which means parameter Q is well estimated. Indeed, values obtained in practice (0.81 for h100, 0.52 for h50 and 0.27 for h10) are consistent with theoretical ones (1.0 for h100).



(a) TNT response (b) EGDN response (b) DCM response

Fig. 4. Example of compounds responses.

Table 1. Recognition rates and times obtained by RT-MSI algorithm.

Explosives	TNT			EGDN		
Concentration	h100	h50	h10	h100	h50	h10
Identification rate	3/3	3/3	3/3	3/3	3/3	3/3
Identification time (s)	47	43	47	31	32	32
Interferents	EtOh			DCM	MEK	
Concentration	h100	h50	h10	h100	h100	
Identification rate	3/3	3/3	2/3	2/3	3/3	
Identification time (s)	35	32	31	31	34	

In Table 2, we have reported results obtained on real data depending on the used technology. When the fluorescence sensor is missing (MBQ+SAW), TNT is not detected anymore while other results are not modified. On the one hand, it outlines the selectivity of the fluorescence sensor which is dedicated to the identification of TNT. Indeed, TNT is the only compound which has a significant response on the OPTO sensor while it does not interact with two other technologies (see Fig : 4). On the other hand, it means that the recognition of EGDN and inteferents is only based on MBQ and SAW sensors. As the responses for these compounds are close (for example, compare EGDN response to DCM one on Fig : 4), performance are deteriorated when the MBQ or SAW sensor is missing (OPTO+SAW or OPTO+MBQ). In these cases, errors have two explanations based on the closeness between models : the system makes confusion between hypothesis or it is not able to take a decision because no compound has its probability becoming greater than 65%. This study outlines the fact that three complementary technologies are necessary to correctly identify and discriminate in real time five classes from which at least four (EGDN and the three interferents) have thin inter-class separations.

Table 2. Recognition rates obtained by RT-MSI algorithm on real data depending on the used technology.

Explosives	TNT			EGDN		
Concentration	h100	h50	h10	h100	h50	h10
OPTO+MBQ+SAW	3/3	3/3	3/3	3/3	3/3	3/3
MBQ+SAW	0/3	0/3	0/3	3/3	3/3	3/3
OPTO+SAW	3/3	3/3	3/3	3/3	2/3	0/3
OPTO+MBQ	3/3	3/3	3/3	3/3	2/3	1/3
Interferents	EtOh			DCM	MEK	
Concentration	h100	h50	h10	h100	h100	
OPTO+MBQ+SAW	3/3	3/3	2/3	2/3	3/3	
MBQ+SAW	3/3	3/3	2/3	2/3	3/3	
OPTO+SAW	2/3	3/3	2/3	2/3	1/3	
OPTO+MBQ	1/3	1/3	2/3	2/3	0/3	

At last, Fig : 5 illustrates the interest in using *a priori* information (regularization and constraints) during the decision step. When values of parameters θ can freely evolve, it appears that all models fit well the real data (Fig : 5.a). In this way, the system cannot afford to take a decision. Constraints on speed evolution and limit values are therefore necessary to assure that only the true hypothesis has its model which correctly fits the real data (Fig : 5.b).

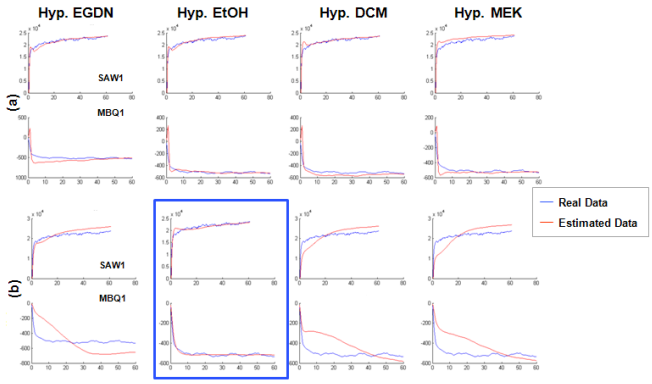


Fig. 5. Illustration of the interest in using constraints with RLS algorithm. The tested compound is EtOh. Only the SAW and MBQ (with the same sensitive material) responses are shown here. On the one hand (a), all models fit well the real data when no constraints are imposed. On the other hand (b), constraints on parameters θ guarantee that only EtOh model fits correctly the real data.

5. CONCLUSION

The RT-MSI approach is a dynamic parameter estimation technique which appears as an alternative to common features extraction methods presented in the literature. Based on the RLS algorithm, the RT-MSI system exploits both kinetics and amplitude ratio between sensors to take an early decision without waiting for the steady state. This ability is the main difference with standard approaches. Classical formulation of the RLS algorithm has been changed to assure the online processing of samples from several sensors with possibly different sampling frequencies. Results obtained on real data outline that our approach is suitable to quickly identify explosive compounds with accuracy.

6. ACKNOWLEDGEMENT

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