

ONLINE MULTITECHNOLOGY SENSORS EXPLOSIVE RECOGNITION

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ABSTRACT

A chemometric method based on an hypothesis test approach built upon RLS (Recursive Least Square) algorithm is presented. This method offers an alternative for online recognition: each analyzed sample leads to the convergence of the system to a state characteristic of the presence of a chemical compound. We present how hypothesis tests are used to exploit this state. In particular we detail some adaptations of RLS used in order to apply our method to a sensor matrix of several technologies in optimal conditions. We show the efficiency of the approach on a real dataset reinforced by statistic results based on simulations.

Introduction

We propose here a signal processing application to explosive compound recognition in the air. We use a solution based on different sensor technologies usually called electronic noses. To exploit these sensors, our system inhales the gas atmosphere to analyze. Then it exposes sensors to the gas.

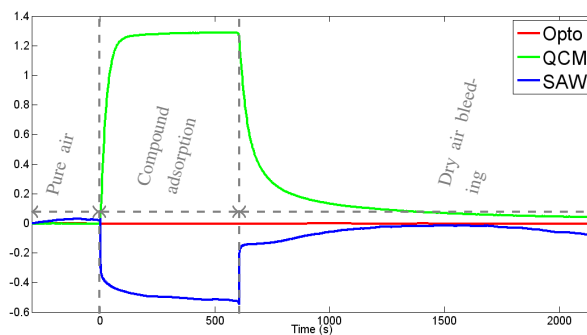


Figure 1: Electronic nose signal acquisition example for TATP exposition. Responses for Fluorescent, Quartz-Cristal Microbalance and Surface Acoustic Wave sensors.

The sensors have a specific response to this exposition due to the gas compound (or vapor) adsorbed by the material coated on its surface. Used technologies are Fluorescent detector (Opto) [2] which leads to intensity variations, Quartz-Cristal Microbalance (QCM) [7] and Surface Acoustic Waves (SAW) [3] which measures frequency variations. These technologies are respectively

either steady and selective but slow, or fast but sensitive to disturbances. As monotecnology system did not give satisfaction for large target objectives, a multitechnology system has been considered. It takes advantage of complementarity of three types of technologies. For the time being each sensor technology correspond to a separate system of which samples are synchronized later on. Then a multitechnological cell will be used. Figure 1 shows some plots of a multisensor example of signals. This case represents triacetone triperoxide (TATP) explosive compound analysis in laboratory conditions. Due to its selectivity to others explosives, fluorescent sensor does not provide response for this gas.

The targets we have to identify are TNT and its impurities (DNT, 4NT), EGDN and oxide water (H_2O_2) as a precursor. We have to estimate the selectivity of the complete system to interferents such as ethanol (EtOH) and methyl ethyl ketone (MEK). In practice, the system will provide two alert levels: first, presence detection of an explosive compound, then its identification. The first alarm exploits sensor selectivity by linear prediction. For second alarm, models of laboratory conditions sensor's responses of each technology are exploited. They can be formulated with sufficient accuracy by Langmuir model [5]:

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

where τ is the characteristic time of the exponential (kinetic), and δ , the sensor sensitivity. These two parameters characterize the interaction between a chemical compound and a sensor. The others parameters define the vector $\theta = (C, \alpha, \beta)^T$. C denotes the compound concentration, α the slope of the sensor linear drift and β the sensor offset. This vector allows us to adjust the model of the presence of a given gas to the real signal.

Over the past few years, several applications of signal processing have exploited the potential of gas sensors for chemical compound recognition. These studies are based on processing of the adsorption part of produced signals. Close to our application, a neural network approach using signal from SnO_2 based on sensors have been proposed by Lee et al. [6]. It allows for example to detect gas such as butane, propane, carbon monoxide and others. Another approach proposed by Bemark et al. [1] combines PCA to Gaussian process models. Both approaches reside in the general framework pre-

sented by Gutierrez-Osuna [4] of learning methods combined with electronic noses. Cited approaches and most of approaches described by Gutierrez-Osuna, as they wait the complete signal acquisition, use end values given only by a monotecnology sensor matrix.

Our approach is an alternative to other chemometric applications as it tackles the lack of learning dataset, drift compensation and calibration transfer (two last objectives described in [4]). We also propose a detection of the adsorption start and a system with fast response to the compound nature. The objective is that average time needed for second alarm is about twenty seconds in spite of compounds' kinetics tested so far. Their average values are around two minutes for fluorescent technology, thirty seconds for QCM technology and ten seconds for SAW technology. The approach is built upon hypothesis tests based on final value and the shape of each signal. Formulated with sensor's response models they are constructed using the linear regression algorithm called Recursive Least Square (RLS).

We present this algorithm and modifications needed to adapt it to our application. Then we present some experiments to real cases and simulated signals.

1. RLS EXPLOITATION

1.1 RLS implementation

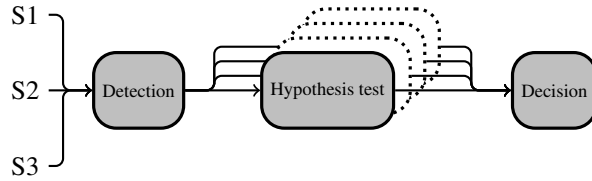


Figure 2: Summary the system architecture: real time detection of the adsorption beginning which activate identification by hypothesis test and decision step.

Detection is based on the localisation of samples passing over a threshold by their distance to the linear drift. Slope of the linear drift and the gap to the drift are estimated for each new sample. Once the beginning of exposition is determined, an identification composed of hypothesis test and decision steps is then applied (steps chronology is illustrated by figure 2). Actually, a one dimensional approach of hypothesis tests with a fusion step to adapt outputs to decision step has first been tested. But gains in signals separability demonstrate the need to use the volatile compound concentration common to all sensors. It leads us to a hypothesis test using intertwined signal. Indeed, it allows to confront sensor sensitivities. Thus, our hypothesis tests built upon RLS algorithm provide a comparison of the adequacy between a model linked to a compound and the signal. Then a decision is taken based on hypothesis tests results. Furthermore, the compound exposition to different sensor is simultaneous. Thus kinetics of each signal can equally be exploited in order to anticipate adsorption final values.

We now remind RLS principle (notations respects those employed in [8]) and we will detail specific modifications applied in order to adapt this algorithm to our issue.

1.2 One dimensional signal case

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

$$Z = H\theta + b, \quad (2)$$

where Z denotes the signal vector, H the model matrix, θ the vector of parameters and b the additive Gaussian noise which depends on the acquisition. θ is coherent with notations of equation (1) defining sensors' models. For a one dimensional signal, one line of the model matrix at index k correspond to $h_k^T = \left(\delta \left(1 - e^{-\frac{t(k)}{\tau}} \right), t(k), 1 \right)$ where $t(k)$ is the instant when the sample z_k is acquired (the sample number k). Parameters which minimize mean square error between signal and model correspond to pseudo-inverse solution $\hat{\theta} = (H^T H)^{-1} H^T Z$. RLS reformulate this pseudo-inverse by updating θ with each new sample in a recursive manner (see [8] for further details):

$$\hat{\theta}_{k+1} = \hat{\theta}_k + P_{k+1} h_{k+1} (z_{k+1} - h_{k+1}^T \hat{\theta}_k) \quad (3)$$

where P_k matrix is an adequate weighting of the different model's parts. Using Sherman-Morrison-Woodbury approach, we can update it by:

$$P_{k+1} = P_k - \frac{P_k h_{k+1} h_{k+1}^T P_k}{1 + h_{k+1}^T P_k h_{k+1}} \quad (4)$$

Multiplied by H_k^T matrix (of the model matrix transposed limited to the analysis support) it provides the pseudo-inverse of H_k .

The main advantage of the method resides in its very low complexity which, for each sample, only depends on the dimension of θ . The more complex step is the update of P_k . The real time application of all assumptions in an embedded system is then achievable.

These notions are the basis of our method. In the following we will list adaptations needed in order to optimize efficiency of the method to our identification framework: multisensor acquisition, regularization on slope parameter α , weighting and concentration positivity constraint.

1.3 Algorithm adaptations

1.3.1 Multisensor adaptation

The most specific aspect of our application is the formulation of the regression for a multisensor system. It is particularly important to formulate the model in order to take into account the common compound concentration. We must also adapt ourselves to channels' acquisitions with their own sampling frequency. We provide here a model

example of an acquisition with two channels. Considering equation (2) and merging all parameters in the same vector, monochannel models become:

$$\begin{aligned} Z_1 &= H_1\theta + b_1 \\ &= \begin{pmatrix} \vdots \\ \delta_1(1 - e^{-\frac{t}{\tau_1}}) & t & 0 & 1 & 0 \\ \vdots \end{pmatrix} \begin{pmatrix} C \\ \alpha_1 \\ \alpha_2 \\ \beta_1 \\ \beta_2 \end{pmatrix} + b_1 \end{aligned} \quad (5)$$

$$\begin{aligned} Z_2 &= H_2\theta + b_2 \\ &= \begin{pmatrix} \vdots \\ \delta_2(1 - e^{-\frac{t}{\tau_2}}) & 0 & t & 0 & 1 \\ \vdots \end{pmatrix} \begin{pmatrix} C \\ \alpha_1 \\ \alpha_2 \\ \beta_1 \\ \beta_2 \end{pmatrix} + b_2 \end{aligned} \quad (6)$$

Equations (5) and (6) allow formulating signals of both sensors acquiring simultaneously the same gas. Our vector θ corresponds to the resolution of the equation implying sample concatenation with $Z_c = [Z_1; Z_2]$ and models concatenation with $H_c = [H_1; H_2]$ (we use here Matlab script notations). In that state, H_c does not provide a real time exploitation of its recursive resolution by RLS algorithm. A reordered version of Z_c and H_c is used instead. In practice, it means that for each sample and according to its original sensor, we build the line of H_c .

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 sensor (the slope and the offset formulated through other columns of the matrix).

1.3.2 Regularization

At the beginning of the regression convergence, slope and exponential evolution are too much correlated to enable pseudo-inverse to discriminate them correctly. We hypothesize that linear drift is low. So in order to avoid confusion, a regularization limiting the slope α (second element of vector θ , see equation 1) has been formulated as:

$$\hat{\theta} = \arg \min_{\theta} (\|Z - H\theta\|^2 + \|\Gamma(\theta - \theta_0)\|^2)$$

The solution proposed by Tikhonov is $\hat{\theta} = (H^T H + \Gamma^T \Gamma)^{-1} H^T (Z - H\theta_0) + \theta_0$ where θ_0 corresponds to initial value of θ . Γ matrix can be defined as the initialization of H given each parameter inertia. Diagonal values of Γ are mainly linked to parameters inertia. This regularization leads to a faster and more robust identification.

1.3.3 Weighting

Weighting samples through the W matrix enables us to adjust the influence of each part of the intertwined signals. Noise level or sampling frequency of each sensor

can for example be adjusted. The estimate equation becomes:

$$\hat{\theta} = (H^T W H + \Gamma^T \Gamma)^{-1} H^T W (Z - H\theta_0) + \theta_0$$

It allows, in our case, to equalize sensors influences by taking into account dynamics of each technology: we define W diagonal terms according to sensor dynamics. Actually, it is equivalent to normalize each technology of sensor according to the maximum value of their outputs.

1.3.4 C parameter positivity

A positivity constraint on the parameter corresponding to the concentration of compounds in gas is also 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.

So a set of constraints around RLS algorithm is available in order to specify hypothesis tests on vector-valued signals. We will then propose a study evaluating the efficiency of this solution according to real and simulated data.

2. RESULTS

As production of real data in laboratory from explosive compound is very complex and hazardous, our validation of the method has been done with a limited corpus of expositions. First we test the solution on real data in order to show the possibility of identification. In order to provide a statistical estimation of the method robustness, we propose some simulations with model variability.

2.1 Real cases

We illustrate the behavior of the system on real data. We start with illustration on two compounds (TATP, TNT) and we show a synthesis on all compounds. Sampling period for both experiences is about one minute for fluorescent, two seconds for QCM and one hundred milliseconds for SAW.

2.1.1 TATP and TNT cases

The TATP exposition presented here corresponds to signal illustrated by figure 1.

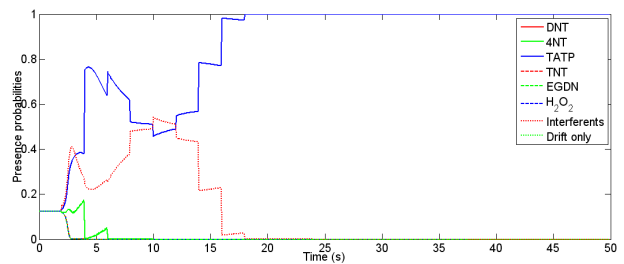


Figure 3: Compounds probability (for TATP exposition).

Figure 3 presents probabilities corresponding to the analysis of this experience. Confrontation of hypothesis tests is presented through probabilities of presence of compounds in gas. The system is able to give the updated probability for each new sample acquired. Thresholds associated to these data give automated decision about explosive compound nature. One is based on the maximum value among probabilities and the other on the ratio between maximum and second maximum among probabilities. This test is an example of fast good identification of TATP. From equiprobability at time zero, an increase of TATP probability quickly leads, after only four seconds, to the decision of its presence. After a light decrease where interferences hypothesis emerge, its probability stabilizes to one. The speed of decision could be very variable like for TNT exposition that we present now.

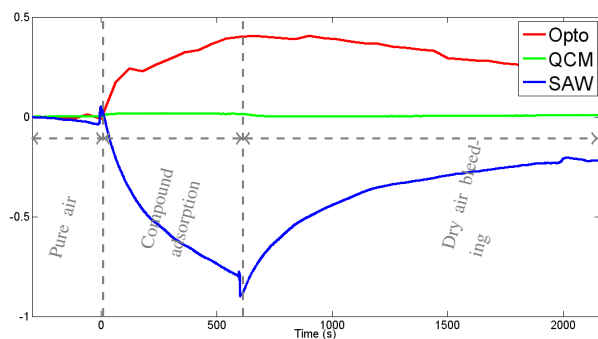


Figure 4: Electronic nose signal acquisition example with an exposition to TNT.

The signal for TNT exposition is presented in figure 4 in the same way we present TATP exposition in figure 1 in order to illustrate their differences.

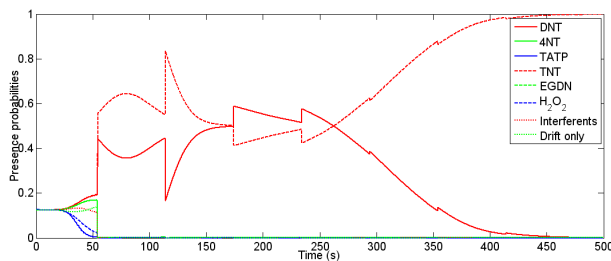


Figure 5: Compounds probability (for TNT exposition).

The evolution of probabilities presented in figure 5 is different to the previous one. Actually, for TNT test (illustrated by figure 5), both DNT and TNT assumptions keep a great probability until 350 seconds. Then TNT hypothesis becomes preponderant. It is due to the similarity of DNT and TNT kinetics and sensibilities. We have to note that this partial confusion is not a problem as both hypothesis lead to the same deduction about explosive presence or not. According to chosen thresholds, the decision is taken for 114 seconds. The probability

locally makes a bounce here as a sample from fluorescent technology arises. Its an illustration of weighting behavior. As fluorescent sample arises every sixty seconds they are given an influence greater than those from QCM and SAW technologies. Both other technologies lead to the regular evolutions of the probabilities. This example shows a slower evolution of probabilities illustrating the influence of similarities in identification behavior for two chemical compounds.

This example also shows the special behavior of the RLS that we propose. Actually each sample updates compounds probability according to its influence. Sampling frequency of fluorescent technology is low. So we use weighting mechanisms in order to give them the same influence in the final decision. Thus, each sample has a great potential influence on probabilities. It is particularly important here as fluorescent technology is selective to TNT but slow. Modifications are applied for future acquisition in order to avoid too much unbalanced sampling frequencies. However, this example is a good illustration of the method flexibility to take into account each technology's characteristics. It should be noted that this method can be adapted to irregular sampling.

2.1.2 Results on a set of tests

We will present here a table summarizing tests results applied with aimed explosives and interferences. Presented results are Δt_d the gap between the real start and the detected start of exposition (localisation reference of the beginning being manual, values are rounded), and Δt_i the identification time when the good compound is designated. These tests correspond to eight experiences of three sensors analyzed on the basis of parameters learned with isolated signals. Kinetics of each compound is around two minutes for fluorescent technology, thirty seconds for QCM technology and around ten seconds for SAW. In this test, we use the detection described in section 1.1.

Table 1: Detection and identification times for each compound on real data are presented here.

	DNT	4NT	TATP	EGDN
Δt_d (s)	0	2	0	0
Δt_i (s)	56	58	4	125
	H_2O_2	TNT	EtOH	MEK
Δt_d (s)	0	6	0	0
Δt_i (s)	3	114	36	24

One can see that detection times reported on table 1 are all respecting objectives. On top of that all identifications for explosives and interferences are correct. Identification times respect objectives of the system as results show times varying from three seconds to two minutes. Variability of these results comes from the distribution of kinetic and sensitivity of sensors linked to tested compounds. As for TATP, H_2O_2 example shows a fast identification: SAW being very selective to it. On the contrary,

like TNT case, some compounds delay the decision making as they have similar responses.

This first approach validates, according to signal acquired until then, the feasibility of the approach. We will now present simulations conceived in order to bench the robustness of identification to model variations.

2.2 Robustness estimation - parameter variations

We present, within the framework of this test, results obtained from multisensor signal simulations based on model described by equation (1). The test is conceived on the basis of a perfect detection in order to focus on identification efficiency. Model variation analyzed here concern adsorption kinetic τ and sensor sensitivity δ . For each model variation we generate one hundred simulations for each tested compound model. A white Gaussian noise is added to simulation with a level learned from real signals. The parameter aimed by the test varies in a uniform manner in an interval according to learnt values. Table 2 indicates ratios of good identification (with their mean times) according to parameter modification.

Table 2: Identification ratios and mean times of good identification presented according to parameter variability.

Modifications	without	$\tau \pm 0.2\tau$	$\tau \pm 0.5\tau$
Id. ratios (%)	100	100	92
Time (s)	17.2	16.8	16.7
Modifications	$\delta \pm 0.2\delta$	$\delta \pm 0.5\delta$	
Id. ratios (%)	99	86	
Time (s)	17.9	17.1	

Results presented by table 2 show a good robustness of the method to kinetics and sensibilities variations. Globally, identification times are stable to changes of these parameters.

This approach provides an estimation of the solution efficiency in respect to parameter variations. It appears from this study that the system is robust to reasonable kinetic and sensitivity changes. Sensibility variations seem to have more influence on identification results than kinetics. We explain these results by the role of sensitivity in gains of multisensor analysis. It is the exploitation of sensors' complementarity through the confrontation of sensitivities that increases discrimination.

Conclusion

We proposed an example of chemical compound recognition in gas based on electronic noses. In order to keep a flexible system (calibration and sensor drift) and based on compounds' adsorption models, an approach by linear regression has been conceived.

Based on RLS algorithm, a real time analysis of samples is proposed with the fastest online answer on the nature of gases in the atmosphere. As this analysis is low in computation cost, it allows a simultaneous application of the method for several assumptions. This method proves to be flexible in physical model definition. Indeed we can

take into account correlations between sensors' behaviors and introduce constraints to the analysis (a priori knowledge) in order to optimize its accuracy to real signals.

Tests with multisensor signals validate the method for real dataset. Confronted to simulated tests signals, results also show a good robustness to kinetic and sensitivity variations.

In the next work we will take aim at a more complex exposition for adaptation from laboratory to real conditions tests.

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