Innovative Procedures to Analyze Data of Gas Sensor Systems and Gas Sensor Nets: a Review

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***Abstract*—Economic online and in-situ field analyses applications like discriminated alarming of smoldering fire or toxic gas leakages, monitoring of volatile components in chemical and biochemical processes, quality monitoring in food processing etc., wait for reliable and economic analytical solutions by sensor systems. This paper presents various innovative mathematical procedures to analyze data from gas sensor systems and gas sensor nets: ProSens - an efficient mathematical procedure for calibration and evaluation of tin oxide gas sensor data, ProCal - a program for batch-wise calibration of multi gas sensors and ProSource - a procedure for source localization. Applications to real data demonstrate the performance of the procedures.**

Keywords: calibration and evaluation procedure; batchwise calibration; source location

I. INTRODUCTION

There is a growing need of economic online and in-situ field analysis applications like online monitoring of volatile components in chemical and biochemical processes, quality monitoring in food processing, discriminated monitoring of toxic gas leakages, etc. In the last application, it is often necessary to find and locate the source of the leakage. In this context, isothermally operated metal oxide gas sensors (MOGs) with tin oxide as base material are manifold introduced due to their high sensitivity, long-term stability and low price. Their sensitivity to specific gas components, however, cannot be cultivated with high discrimination to others. Therefore, other approaches are necessary like a multi gas sensor array of MOGs [1][2] or thermo-cyclic operation of the MOG.

For evaluation of the sensor data, powerful mathematical evaluation procedures for substance identification and concentration determination, even in the case of variable environmental conditions like varying humidity, are necessary. The calibration of sensor elements is very time consuming and expensive. Even sensor elements which are fabricated batch-wise have to be individually calibrated for good analysis performance. Therefore, economic mathematical calibration procedures are useful to reduce the costs and the scope of calibration measurements. Last but not least, mathematical procedures are needed to locate the source of a leakage.

At the Karlsruhe Institute of Technology (KIT) mathematical procedures are developed to meet the above mentioned requirements: ProSens – a mathematical procedure for calibration and evaluation, ProCal – a mathematical procedure for batch-wise calibration of sensor elements and ProSource – a mathematical procedure to locate the source of a leakage. After some remarks to thermo-cyclic operation of the MOG, these procedures will be briefly described in this paper and their performance will be demonstrated in applications with real data.

In Section 2, the principles of thermo-cyclic operation are briefly discussed. A short outline of the program ProSens with two applications to real data is given in Section 3. In Section 4, the program ProCal is briefly introduced and its performance is demonstrated in an example. In Section 5, the program ProSource for source localization is briefly described and in Section 6, the results of the paper are summarized.

II. THERMO-CYCLIC OPERATION

Thermo-cyclic operation means, that the working temperature of the sensor element is periodically increased and decreased over the time in a triangular shape. Simultaneous sampling of the conductance values over the time leads to so-called Conductance over Time Profiles (CTP) [3][4][5]. These profiles give a fingerprint of the surface processes with the gas and represent the gas mixture under consideration. The gas specific features of the CTPs can be used for component identification and concentration determination.

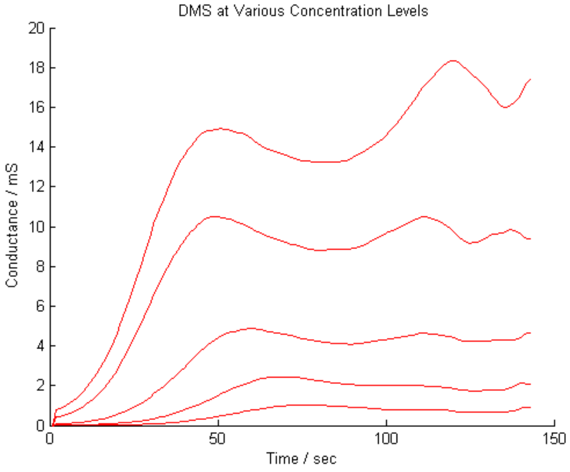

Figure 1. CTPs of dimethyl sulphate (DMS) at various concentration levels

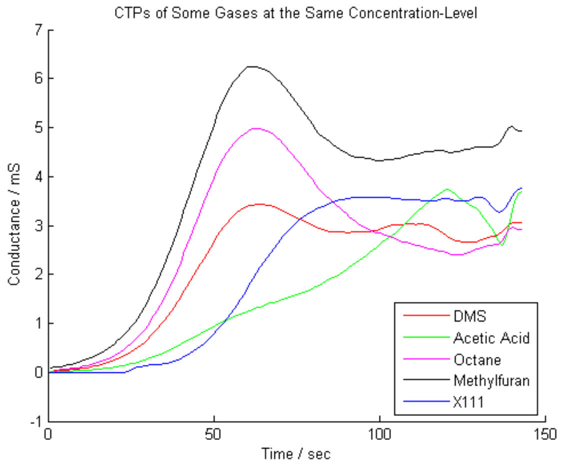

Figure 2. CTPs of some gases at a certain concentration level

Figure 1 shows the CTPs of dimethyl sulphate (DMS) at various concentration levels with the characteristic shape. The working temperature of the sensor element is in the range of 100 to 400 degree Celsius. The conductance values are recorded only over the growing part of the triangular shape.

Figure 2 shows the CTPs of some gases at a certain concentration level. It can be seen that the shapes of the various gases are quite different. Therefore, they can be used for substance identification using innovative calibration and evaluation procedures.

III. PROSENS – A MATHEMATICAL PROCEDURE FOR CALIBRATION AND EVALUATION

As mentioned above, MOGs can be used for ambitious analysis applications, if they are thermo-cyclically operated. A mathematical procedure for data evaluation has to identify an unknown gas sample (classification) to avoid misleading results like false alarms. Furthermore, it has to determine the concentration of the components of the gas sample. Varying environmental conditions like varying humidity or varying environmental temperature often influence the measurement results. These variations must be incorporated in the evaluation model of the mathematical procedure.

The program ProSens developed at KIT was designed to meet the above mentioned requirements. ProSens consists of two parts: a calibration part and an evaluation part.

In the calibration part, ProSens provides the mathematical calibration model which is the basis for routine data evaluation. The determination of the mathematical calibration model is based on calibration measurements performed by MOG with thermo-cyclic operation. These mathematical calibration models are parametric models. Their parameters are transferred to the evaluation part for the analysis of unknown gas samples.

In the evaluation part of ProSens, an unknown gas sample is analyzed. That means that a substance identification and a concentration determination is performed.

For gas component or compound identification, ProSens calculates the so-called theoretical CTP for the calibrated gas under consideration and compares this CTP with the measured CTP.

If the measured CTP and the theoretical CTP are close together, i.e., a difference value calculated from the sum of quadratic differences of every sample point of the measured CTP and the theoretical CTP is smaller than a predetermined decision value, ProSens identifies the unknown gas sample with the related calibrated gas.

Otherwise ProSens recognizes that the gas sample is not the calibrated gas.

In the case of a successful identification, ProSens calculates the concentration of the gas sample.

In order to demonstrate the performance of the mathematical evaluation procedure ProSens, the CTP-data of two evaluation experiments of chemical analysis were used, first ammonia under various water vapor partial pressures and second binary toluene/ethanol-mixtures dissolved in water. The experimental details of data acquisition are described in [5][6].

In the first investigation ammonia was analyzed. To perform the mathematical calibration model the CTP was measured at different ammonia concentrations (100 ppm, 200 ppm, 500 ppm, 1000 ppm) and at different water vapour partial pressures (529, 1322, 2334 Pa) as well. These 12 calibration points are visualized in Figure 3. This low number of calibration points is important, because it keeps the calibration costs low. The graphs of measured and theoretical CTP are compared when testing an ammonia sample (750 ppm) in Figure 4(a) and in case of a non-ammonia sample (carbon monoxide with 1000 ppm) in Figure 4(b). It is evident that ProSens is able to identify the ammonia sample. Table I demonstrates that ProSens can determine the concentration of ammonia samples quite well, even in the case of varying water vapour partial pressures.

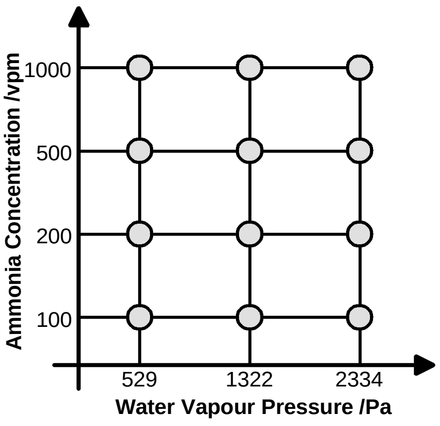

Figure 3. Calibration field of only 12 calibration points

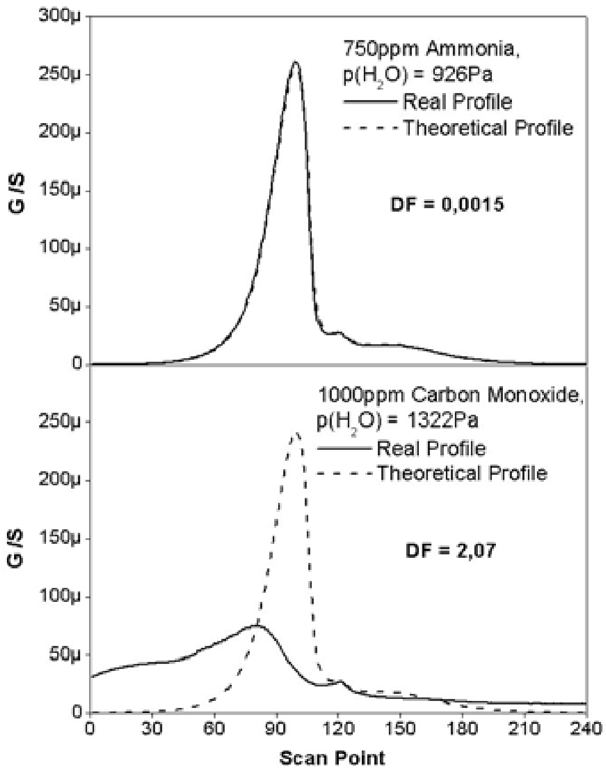

Figure 4. Comparison of measured/theoretical CTP (a) ammonia sample, (b) non-ammonia sample

TABLE I. AMMONIA ANALYSIS RESULTS OF TEST EXPERIMENTS AT DIFFERENT WATER PARTIAL PRESSURES

Dosed concentration	p(H2O)/Pa	Determined concentration
150 ppm	1058	159 ppm
150 ppm	1587	171 ppm
350 ppm	1058	329 ppm
350 ppm	1587	373 ppm

In a second example, the CTP-data of binary toluene/ethanol mixtures dissolved in water were used for chemical analysis. Also in this case only 12 calibration points were used in the calibration procedure (Figure 5). Three further binary toluene/ethanol-mixtures (analysis test points, Figure 5) and a non-toluene/ethanol-mixture were analyzed for demonstration of the performance of ProSens.

The analysis proves that the evaluation procedure is able to identify toluene/ethanol binary mixtures and to detect non-toluene/ethanol-mixtures as well. This is due to the fact that in the second case the difference between the theoretical CTP and the measured CTP is significantly big (Figure 6) in comparison with the negligible difference in the first case (Figure 7).

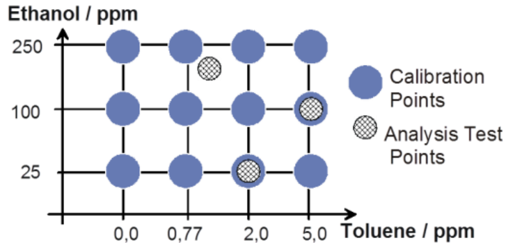

Figure 5. Calibration field and analysis test points of toluene/ethanol mixtures dissolved in water at constant temperature. Sensor signals were sampled using a gas carrier sample.

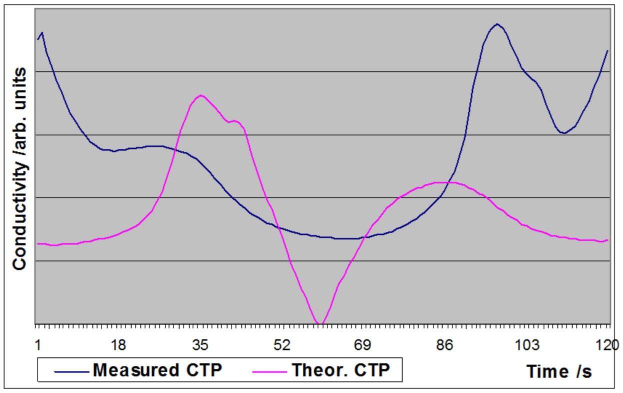

Figure 6. Comparison of measured and theoretical CTP of a non-toluene/ethanol-mixture

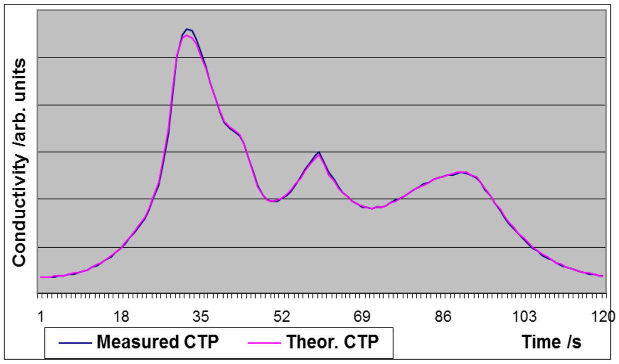

Figure 7. Comparison of measured and theoretical CTP of a toluene/ethanol binary mixture

TABLE II. RESULTS OF THE ANALYSIS TESTS

Dosed concentrations	Determined concentrations	Time delay*/days
25/2	23.4/2.1	1.25
100/5	95.3/5.4	1.0
200/1	200.1/0.9	1.5

*Time delay of the analysis test experiment after having finished the calibration data sampling

The CTP-analysis resulted in rather accurate estimations of the VOC-components (error < 10%), see Table II.

IV. ProCal – A Mathematical Procedure for Batch-wise Calibration

For economic reasons, MOGs are typically fabricated in batch production. For technological reasons, production fluctuations are unavoidable which lead to fluctuations of the gas sensors specific features, e.g., manifested in CTP shape variations in the case of thermo-cyclic sensor operation.

Although the signal patterns (e.g., CTPs) of the various sensor chips of a production line are similar, each sensor chip has to be costly calibrated, in order to yield a high analytic performance. The same is true if a sensor chip has to be exchanged in case of a defect or if a sensor chip has to be recalibrated in case of long-term drifting.

The basic idea of ProCal is that only one single sensor chip (the so-called class reference chip) of the production line is measured at all calibration points (typically 5 calibration points for a single gas analysis application) and its signal patterns are sampled. Every other chips of the production line are measured at only one calibration point (the so-called reference point).

Next, for every sensor chip a mathematical function (the so-called approximation function) is determined which fits the signal pattern of the reference point, yield by the reference chip, to the signal pattern yield by the corresponding sensor chip in a best manner. That means that for each sensor chip excluding the reference chip a sensor specific approximation function is obtained.

With these approximation functions, the signal patterns of the reference chip are numerically calculated for all the other calibration points of the corresponding chips of the batch. This means that the signal patterns for the other calibration points do not have to be measured but can be approximated by numerical calculation. This results in a significant reduction of the calibration measurements by approximately factor 5.

The whole procedure is sketched in Table III with sensor s1 as the reference chip and concentration con3 as the reference point.

Furthermore, ProCal is able to find the “optimal” reference chip and to recognize a priori which sensor chips of the production line cannot be calibrated with this procedure.

To demonstrate the effectiveness of ProCal, an application was performed with eight thermo-cyclically operated gas sensors which were exposed to 0 ppm, 250 ppm, 500 ppm, 1000 ppm and 2000 ppm CO in humidified synthetic air (50% rH, 21°C), as described in [7].

In ProCal, only the CTPs of the reference concentration point (here at 500 ppm CO) of every sensor (Figure 8) are used to calculate sensor specific approximation functions.

ProCal recognizes that sensor S4 is the best choice for the class-reference chip.

Furthermore, ProCal a priori determines sensor S3 as an outlier which cannot be calibrated with this procedure.

TABLE III. SCHEME OF THE CALIBRATION PROCEDURE

	s1	si	s1: class reference chip
con1	m	n	si = 2,3,..(other sensors)
con2	m	n	m:meas. signal pattern
con3	m →	m	→:approx. function f_i
con4	m	n	n: num. calcul. with f_i
con5	m	n	con3: ref. calibr. point

f_i:CTP(s1,con3) -> CTP(si,con3) “optimal”

f_i: CTP(s1,conj) -> CTP(si,conj), j = 1,..,5

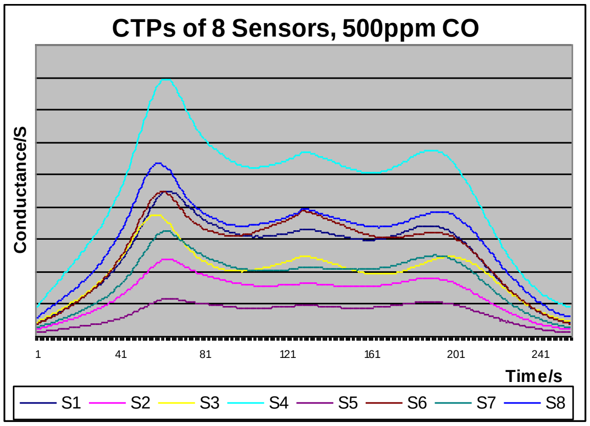

Figure 8. CTPs of eight sensors at reference point 500ppm CO. Sx means sensor x

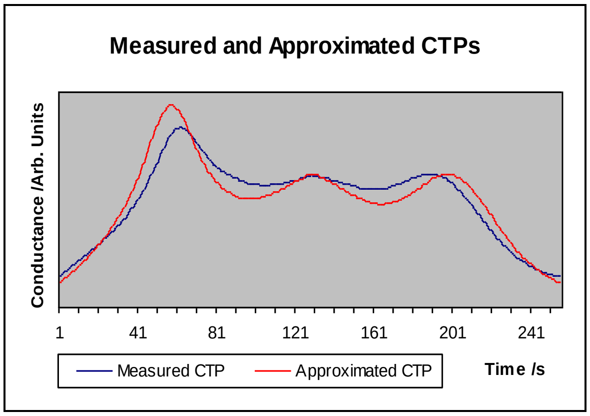

Figure 9. Measured and approximated CTP of S3 at reference point 500 ppm CO

Indeed, Figure 9 shows that the difference between numerically calculated CTP (using the relevant approximation function) and the sampled CTP is strikingly big for sensor S3 at reference point 500 ppm CO.

To demonstrate how well the numerically measured CTP fits the sampled CTP, the data of S2 at 1000 ppm CO are visualized in Figure 10. Similar results are obtained at all calibration points of the other sensors except the a priori excluded sensor S3.

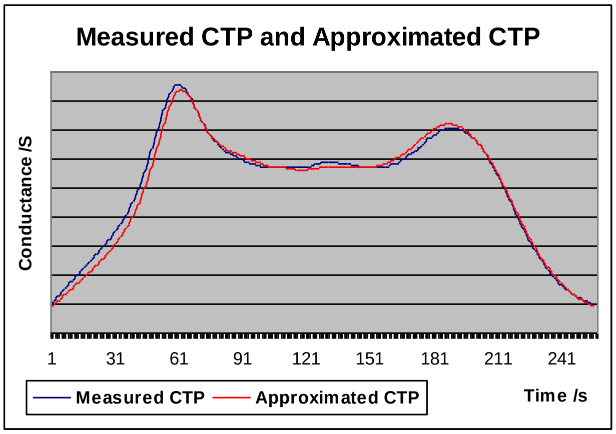

Figure 10. Comparison of measured CTP and approximated CTP of sensor S2 at exposure of 1000ppm CO

TABLE IV. ANALYSIS RESULTS IN PPM CO

DC	S1	S2	S3	S5	S6	S7	S8
0	13	11	-4	5	10	15	2
250	258	250	205	234	247	262	240
500	495	495	495	495	495	495	495
1000	948	988	1180	1007	1014	951	1004
2000	1801	1984	2559	1943	1977	1788	1937
MD	9.95	1.2	27.9	6.4	1.4	10.6	4.0

*DC: Dosed concentration, MD: Maximum deviation from DC in % (DC≠0)

For the determination of the mathematical calibration model with ProSens, only the measured CTPs at the reference point 500ppm were used. All the other CTPs needed for the determination of the calibration model were not measured but calculated. Table IV shows the analysis results achieved by ProSens. The relative analysis errors in all considered cases are less than 11%, except for the a priori excluded sensor S3. These errors are comparable with those obtained when using sampled data.

V. PROSOURCE - A PROCEDURE FOR SOURCE LOCALIZATION

Based on concentration measurements from spatially distributed gas sensors s_i (gas sensor network), the location of a gas source (e.g., from a leakage) is to be determined. To solve this problem it is assumed that the emitted substance is transported by advection caused by a known homogeneous wind field in x-direction and by diffusion.

The basic idea of the procedure ProSource [8][9] is to use a two-step approach for the source localization.

In the first step, for each sensor s_i, located at x_i, the set of points P_i is determined, on which the source can lie, taking only the specific concentration measurement C_i of sensor s_i into account.

In the second step, an estimate for the source position x_0 is evaluated by intersecting the sets P_i.

A. *Determining P_i*

Each set P_i is given by:

$$\mathcal{P}_i = \left\{\mathbf{x}_0 \in \mathbb{R}^2 : \|\mathbf{x}_i - \mathbf{x}_0\|_2 = d_i(C_i, x_0, q_0)\right\} \quad (1)$$

Here, for a fixed but unknown source rate q_0 and the measured concentration C_i, the set P_i forms an oval in the x-direction. For different assumed source rates q_0 the oval sets P_i for one sensor s_i are plotted in Figure 11.

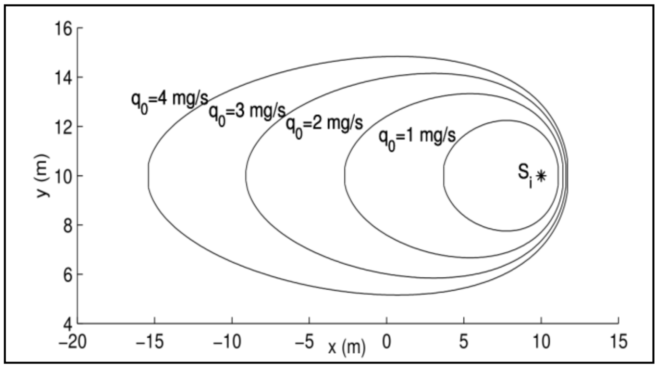

Figure 11. Potential source positions for sensor s_i for different source rates q_0

B. *Estimating the source position x_0*

Like for one sensor in Figure 11, ovals can be calculated for all m sensors of the sensor network. Thus, all m ovals intersect at the source position x_0, if the source rate, used to generate the ovals, equals the true value q_0. This is demonstrated in Figure 12. In this case, four gas sensors are applied.

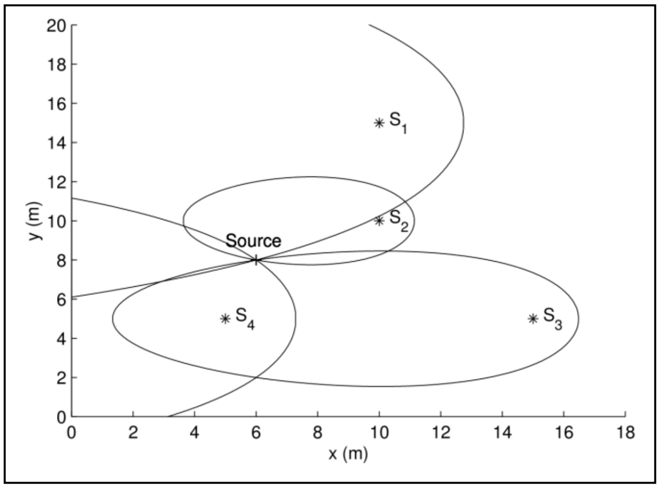

Figure 12. Intersection of all m sets P_i at the source position x_0 for the undisturbed case and the true value of q_0

Figure 12 demonstrates that in the undisturbed case the intersection of the curves is the source position of the gas leakage with the corresponding source rate. In the disturbed case, ProSource calculates an estimated source position and a corresponding estimated source rate that minimize the geometric error of all ovals.

VI. CONCLUSIONS

In this paper the mathematical procedures ProSens, ProCal and ProSource have been shortly described and their performances have been demonstrated with examples using real measurement data.

The examples demonstrate that the procedures developed at KIT are powerful tools for economic online and in-situ field analyses applications. ProSens is an efficient mathematical procedure for evaluation of tin oxide gas sensor data, even in the case of variable environmental conditions like varying humidity. ProCal is an economic mathematical procedure for batch-wise calibration of multi gas sensors, useful to reduce the costs and the scope of calibration measurements. ProSource is a robust and fast procedure for source localization based on spatially distributed concentration measurements.

ProSens is designed to analyze only one gas mixture under consideration. But in many applications a multitude of different gases may occur, for example in detecting cable fire in a cable slot at a very early point of time but with high discrimination to other gases. Depending on the various coating materials of the cables, a variety of gases or gas mixtures can be emitted which have to be simultaneously identified and analyzed for early detection of such a developing risk. For this reason, ProSens has to be extended in a next step to meet these requirements.

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