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Mobile Sensor System AGaMon for Breath Control: Numerical Signal Analysis of Ternary Gas Mixtures and First Field Tests

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Abstract: An innovative mobile sensor system for breath control in the exhaled air is introduced. In this paper, the application of alcohol control in the exhaled air is considered. This sensor system operates semiconducting gas sensor elements with respect to the application in a thermo-cyclic operation mode. This operation mode leads to so-called conductance-over-time-profiles (CTPs), which are fingerprints of the gas mixture under consideration and can be used for substance identification and concentration determination. Especially for the alcohol control in the exhaled air, ethanol is the leading gas component to be investigated. But, there are also other interfering gas components in the exhaled air, like H2 and acetone, which may influence the measurement results. Therefore, a ternary ethanol-H2-acetone gas mixture was investigated. The establishing of the mathematical calibration model and the data analysis was performed with a newly developed innovative calibration and evaluation procedure called ProSens 3.0. The analysis of ternary ethanol-H2-acetone gas samples with ProSens 3.0 shows a very good substance identification performance and a very good concentration determination of the leading ethanol component. The relative analysis errors for the leading component ethanol were in all considered samples less than 9 %. First field test performed with the sensor system AGaMon shows very promising results.

Keywords: Alcohol control, Mobile sensor system, Thermo-cyclic operation, Data analysis, Substance identification, Concentration determination, Field test.

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1. Introduction

There is a broad field of applications of breath monitoring in human health care, medical applications and alcohol control. In this context, several approaches are suggested [1]. In particular, metal oxide gas sensors (MOG) can be used as appropriate candidates for breath control. This is due to the fact that they are very sensitive, have good long-term stability and are low in price.

But, on the other hand, when these sensor devices are operated isothermally, they are not at all selective. That means that they cannot be used for sophisticated analysis of gas mixtures. Therefore, other approaches are necessary like a gas sensor array of MOGs [2-3] or by thermo-cyclic operation of the MOG and simultaneous sampling of the conductance, which leads to so-called “conduction over time profiles” (CTPs) [4-7].

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. At the Karlsruhe Institute of Technology (KIT), many procedures were established to evaluate such signal patterns [8], for batch-wise calibration of sensor elements [9] and also for source localization [10].

In this report [11], an innovative mobile sensor system AGaMon (AtemGasMonotor, Breath Control Monitor) for breath control in the exhaled air is introduced. Especially for alcohol control, which is the investigated application in this paper, ethanol is the leading component.

But, because other components like H2 and acetone may also occur in the breathing air and may influence the measurement results, ternary ethanol-H2-acetone gas mixtures are considered. This is an important update to the investigation performed in [12]. The analysis of these samples is performed with the calibration and evaluation program for ProSens 3.0, which is an integral component of the sensor system.

In Section 2, the mobile sensor system AGaMon is described. A short outline of the calibration and evaluation procedure ProSens 3.0 is given in Section 3. In Section 4, the data analysis for the application alcohol control with ternary ethanol-H_2-acetone gas mixtures is given, including the calibration set up, substance identification and concentration determination of the leading component ethanol. In Section 5, the results of a first field test of the sensor system AGaMon are given. Section 6 summarizes the results of this paper.

2. Mobile Sensor System AGaMon

2.1. Sensor System Platform and Adapter

For breath control in the air we exhale, especially for alcohol control, an innovative sensor system platform AGaMon was developed. Based on this platform, an adapter for smartphones was developed for mobile monitoring of the breathing air.

This adapter consists of a combined and modular hardware- and software system, which runs an embedded metal oxide gas sensor in a thermo-cyclic mode and which determines the alcohol content on the basis of the measurement results via an innovative calibration- and evaluation procedure ProSens 3.0 in real time. The analysis results will then be displayed on the smartphone.

The following Fig. 1 shows a pre-release version of the mobile sensor system.

2.2. Electronics for Heater Control and Data Acquisition

In order to characterize and operate semiconducting gas sensor elements with respect to the application, a sensor platform was developed, which ensures a robust functioning of hard- and firmware. This platform supports a variety of commercially available metal oxide gas sensors. In this investigation, the sensor MLV (MultiLayer Varistor) from Applied Sensors [13] was used. Via its graphical user interface, different parametriseable temperature cycles can be configured.

Fig. 1. Pre-release Version of the Mobile Sensor System.

Additionally, this system allows the sensors to be exposed to several interfering gases like: H2S (which is the leading component for halitosis), H2 (which is the leading component for dyspepsia and food intolerance), NO (which is the leading component for asthma) or Acetone (which is the leading component for diabetes), thus covering almost all significant aspects.

The core unit of the platform is a base-board with a powerful micro-controller communicating with external modules in a master-slave-configuration. The base-board is able to manage up to four gas sensor modules and features ambient condition monitoring.

The platform outputs the sensor raw data (basically the measured voltages), which can easily be transformed into resistances or conductances or pre-calculated values for a reduced data stream. Via USB, the platform is connected to a standard PC where the data live visualization and the storage is carried out. Via Bluetooth, the platform can be connected to mobile applications running on smart phones.

For the measurements in this paper, a platform with the following specifications was used:

- The temperature control allows a set-point accuracy of 2 °C within an overall temperature range of 100 to 500 °C. The set-point can be updated every 10 ms.
- The read-out circuit features a sampling time of better than 1 ms.
- Measurement voltage accuracy is around of 5 mV (by using a 10-bit-ADC).
- The dynamic range of the read-out circuit is between 1 k and 100 M.

2.3. Temperature Cycle

Based on the above-explained electronics, several temperature cycles have been applied to the sensors while being exposed to the gas mixtures.

For the experiments carried out in the scope of the publication, the temperature cycle in Fig. 2 has been considered. It could be shown that this temperature cycle provides the best analysis results regarding the application under consideration.

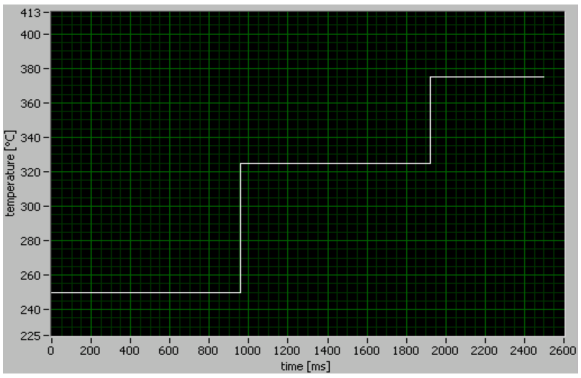

Fig. 2. Thermo-cyclic (step-wise) temperature cycle.

3. Calibration and Evaluation Procedure ProSens 3.0

As mentioned above, the calibration- and evaluation procedure ProSens 3.0 is included as an integral component in the mobile sensor system. ProSens 3.0 is an updated version of ProSens [14] to meet the requirements of this sensor system and to analyze ternary gas mixtures. Similar to ProSens, ProSens 3.0 consists of a calibration part and an evaluation part.

Using the calibration part of ProSens 3.0, the mathematical calibration model is calculated based on calibration measurements. The mathematical calibration model is a parametric model and only the parameters will be transferred to the evaluation part of ProSens 3.0. This is very important because all the time consuming calculations can be performed off-line.

If an unknown gas sample is measured, the evaluation part of ProSens 3.0 performs a substance identification and concentration determination of the sample, based on the calibration parameters. For substance identification, ProSens 3.0 determines a calculated CTP and compares this CTP with the real measured CTP. Only if the distance of calculated CTP and measured CTP is smaller than a pre-determined decision threshold, ProSens 3.0 identifies the unknown sample with the gas sample under consideration. In this case, the concentration determination will be performed.

Substance identification is very important to avoid misleading analysis results like false alarms.

4. Application – Alcohol Control in the Exhaled Air

As mentioned in Section 2.1 and Section 2.2, the mobile sensor system is suitable for a broad range of applications for breath monitoring.

In this application, we turn the focus to the investigation of the alcohol control in the exhaled air. In this context, ethanol is the leading component. But, there are also other interfering gas components in the air we exhale, like H_2 and acetone, which may influence the measurement results. Therefore, ternary ethanol-H2-acetone gas mixtures are investigated.

The measurements were performed with the sensor system described in Section 2 using the cyclic variation of the working temperature in Fig. 2. The determination of the mathematical calibration model and the data analysis were performed with the included program ProSens 3.0.

4.1. Calibration Set Up

In order to establish the mathematical calibration model using the calibration part of ProSens 3.0, calibration measurements with dosed concentrations of the ternary gas mixtures have to be performed. The following Table 1 shows the concentrations of the gas mixtures, which are used for calibration.

Table 1. Gas Samples used for Calibration.

Ethanol-H_2-Aceton in ppm	**Ethanol-H_2-Aceton in ppm**	**Ethanol-H_2-Aceton in ppm**
50-10-0.5	50-10-1.0	50-10-2.0
100-10-0.5	100-10-1.0	100-10-2.0
175-10-0.5	175-10-1.0	175-10-2.0
50-20-0.5	50-20-1.0	50-20-2.0
100-20-0.5	100-20-1.0	100-20-2.0
175-20-0.5	175-20-1.0	175-20-2.0
50-30-0.5	50-30-1.0	50-30-2.0
100-30-0.5	100-30-1.0	100-30-2.0
175-30-0.5	175-30-1.0	175-30-2.0

It can be seen that only 27 samples were used for establishing the mathematical calibration model for the ternary mixture. This is a very good result, because calibration measurements are very time-consuming and expensive.

4.2. Data Analysis

To investigate the performance of the sensor system with the evaluation procedure ProSens 3.0, 9 further ternary ethanol-H_2-acetone gas mixtures and a foreign substance were measured in the same manner as the samples for calibration and analyzed together with the samples of the calibration process. The samples are given in Table 2.

The red marked samples are additionally measured samples. The foreign substance is not listed in Table 2.

Table 2. Samples used for Evaluation.

Ethanol-H_2-Aceton in ppm	Ethanol-H_2-Aceton in ppm	Ethanol-H_2-Aceton in ppm	Ethanol-H_2-Aceton in ppm
50-10-0.5	175-10-0.5	**135-20-0.5**	100-30-0.5
50-10-1.0	175-10-1.0	**135-20-1.0**	100-30-1.0
50-10-2.0	175-10-2.0	**135-20-2.0**	100-30-2.0
100-10-0.5	50-20-0.5	175-20-0.5	**135-30-0.5**
100-10-1.0	50-20-1.0	175-20-1.0	**135-30-1.0**
100-10-2.0	50-20-2.0	175-20-2.0	**135-30-2.0**
135-10-0.5	100-20-0.5	50-30-0.5	175-30-0.5
135-10-1.0	100-20-1.0	50-30-1.0	175-30-1.0
135-10-2.0	100-20-2.0	50-30-2.0	175-30-2.0

4.3. Substance Identification

For substance identification, as already mentioned in Section 3, the calibration and evaluation ProSens 3.0 calculates the so-called calculated CTP and compares this CTP with the real measured CTP.

Fig. 3, Fig. 4 and Fig. 5 give a visual impression of calculated CTPs and measured CTPs.

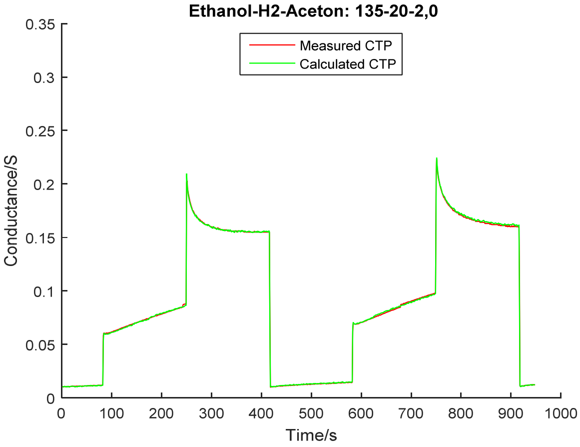

Fig. 3. Comparison of measured CTP and calculated CTP: Ethanol 135 ppm/H_2 20 ppm/Acetone 2 ppm.

In Fig. 3 and Fig. 4, the calculated CTP (green line) and measured CTP (red line) of ternary ethanol-H_2-acetone samples are plotted. It can be clearly seen, that the difference between the two curves is in both cases very small. This means that ProSens 3.0 recognizes that these samples are the ternary gas mixtures under consideration.

Theoretical CTP and measured CTP for the foreign substance are shown in Fig. 5. In this case, the calculated CTP is not so close to the measured CTP as in the case of the ternary ethanol-H_2-acetone gas mixtures. So, the difference between the two curves is much larger. That means that ProSens 3.0 recognizes that this sample is not the calibrated ternary gas mixture.

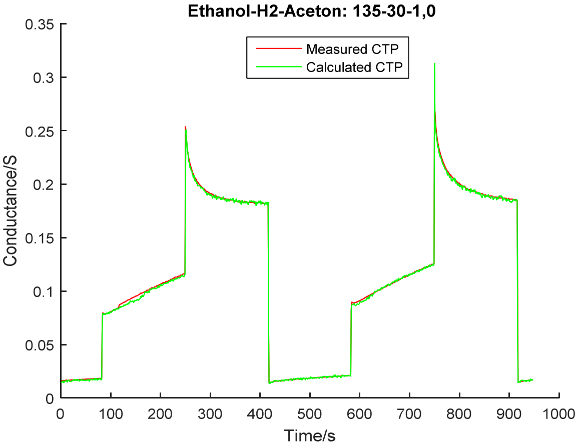

Fig. 4. Comparison of measured CTP and calculated CTP: Ethanol 135 ppm/H_2 30 ppm/Acetone 1 ppm.

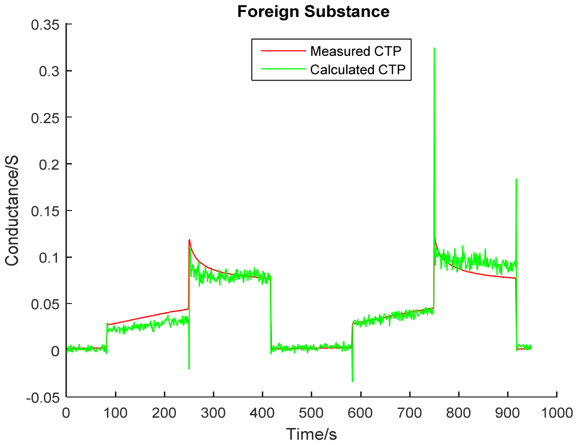

Fig. 5. Comparison of measured CTP and calculated CTP of a Foreign Substance.

Of course, the decision for substance identification is not based on the visual impression. Therefore, a "difference value" is calculated from the sum of quadratic differences of every sample point of the measured CTP and the theoretical CTP. Only if this difference value is smaller than a predetermined decision value, ProSens 3.0 identifies the unknown gas sample with the related calibrated gas mixture. Table 3 shows the difference value for the gas samples.

In Table 3, the green highlighted rows correspond to the difference values according to the ternary ethanol-H_2-acetone mixtures, the dark green rows to ternary samples additionally measured for evaluation and the red row to the foreign substance. The difference value according to the foreign substance is 0.57 and much larger than the difference values according to the ternary gas mixtures, which are in all considered cases equal or smaller than 0.0035.

Therefore, it is evident that the difference values in the green rows are smaller in dimensions than the difference value in the red row.

Table 3. Difference Values (Between Measured CTP and Calculated CTP).

Ethanol/H2 (*Aceton=0.5 ppm)*	**10 ppm**	**20 ppm**	**30 ppm**
50 ppm	0.0000	0.0001	0.0001
100 ppm	0.0001	0.0002	0.0001
135 ppm	0.0000	0.0041	0.0001
175 ppm	0.0000	0.0000	0.0000
Ethanol/H2 (*Aceton=1.0 ppm)*	**10 ppm**	**20 ppm**	**30 ppm**
50 ppm	0.0003	0.0000	0.0001
100 ppm	0.0000	0.0000	0.0000
135 ppm	0.0003	0.0027	0.0133
175 ppm	0.0003	0.0000	0.0001
Ethanol/H2 (*Aceton=2.0 ppm)*	**10 ppm**	**20 ppm**	**30 ppm**
50 ppm	0.0001	0.0001	0.0001
100 ppm	0.0000	0.0004	0.0000
135 ppm	0.0022	0.0022	0.0035
175 ppm	0.0001	0.0000	0.0000
Foreign Substance	0.5706		

If the decision value is set, for example, to 0.1, there is good discrimination between the difference values of the ternary gas mixtures under consideration and the difference value of the foreign substance. That means that ProSens 3.0 is able to perform a very good substance identification.

4.4. Concentration Determination

After substance identification, ProSens 3.0 performs the concentration determination of the gas samples, which were identified as the ternary ethanol-H2-acetone gas mixtures. In the application under consideration, ethanol is the leading component. That means that only the concentrations of the ethanol components of the ternary mixtures are essential. The following Table 4 shows the calculated concentrations of the ethanol component in comparison to the dosed values of the ternary ethanol-H2-acetone gas mixtures.

Next, Table 5 shows the relative analysis errors for the estimation of the ethanol concentration of the ternary gas mixture.

It can be seen that, in all cases, the relative analysis error for the ethanol concentration is smaller than 9 %. This is a very good analysis result.

Table 4. Analyzed Ethanol Concentration in ppm.

Ethanol/H2 (*Aceton=0.5 ppm)*	**10 ppm**	**20 ppm**	**30 ppm**
50 ppm	49.6	49.1	51.2
100 ppm	100.3	102.7	100.1
135 ppm	**131.5**	**141.0**	**133.8**
175 ppm	177.0	177.7	176.2
Ethanol/H2 (*Aceton=1.0 ppm)*	**10 ppm**	**20 ppm**	**30 ppm**
50 ppm	50.6	50.5	50.0
100 ppm	102.1	99.0	99.8
135 ppm	**126.4**	**139.0**	**142.1**
175 ppm	174.7	174.0	172.7
Ethanol/H2 (*Aceton=2.0 ppm)*	**10 ppm**	**20 ppm**	**30 ppm**
50 ppm	49.7	49.9	49.0
100 ppm	98.0	98.2	100.3
135 ppm	**128.0**	**136.2**	**123.5**
175 ppm	173.1	173.1	176.0

Table 5. Relative Analysis Errors for the Ethanol Deamination in %.

Ethanol/H2 (*Aceton=0.5 ppm)*	**10 ppm**	**20 ppm**	**30 ppm**
50 ppm	0.8	1.7	2.4
100 ppm	0.2	2.7	0.0
135 ppm	**2.6**	**4.5**	**0.9**
175 ppm	1.1	1.6	0.7
Ethanol/H2 (*Aceton=1.0 ppm)*	**10 ppm**	**20 ppm**	**30 ppm**
50 ppm	1.2	1.0	0.0
100 ppm	2.1	1.0	0.2
135 ppm	**6.4**	**2.9**	**5.3**
175 ppm	0.2	0.6	1.3
Ethanol/H2 (*Aceton=2.0 ppm)*	**10 ppm**	**20 ppm**	**30 ppm**
50 ppm	0.6	0.2	1.9
100 ppm	2.0	1.8	0.3
135 ppm	**5.2**	**0.9**	**8.5**
175 ppm	1.1	1.1	0.6

5. First Field Tests

The above obtained very good analysis results are based on measurements in the laboratory. To prove the performance of the system not only to laboratory data, the sensor system was applied to first field tests.

In this field test, 6 persons were involved as test persons, in the following called probands.

The probands trunk during the test period dosed amounts of alcohol in 6 fixed time intervals. After each time interval, there was a waiting time of 15 minutes before the measurements were performed.

After each waiting time, the measurements of the blood alcohol concentration were first performed by a reference measurement device, then by the sensor system AGaMon.

In the following Fig. 6, the measurement results of every proband, obtained by the sensor system AGaMon, are displayed against the measurement results obtained by the reference measurement device.

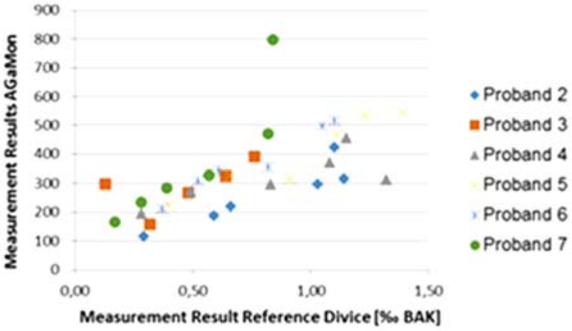

Fig. 6. Comparison of Measurement Results obtained by the AGaMon System against the Measurement Results obtained by the Reference Device for every Proband.

It can be clearly seen in Fig. 6 that there is a strong correlation between the measurement results obtained by the sensor system AGaMon and the measurement results obtained by the reference measurement device. This is a very promising result. Nevertheless, it can be also seen that there are some outliers in the measurement results of AGaMon. This can be due to some technical problems or due to erroneous handling of the proband.

In the following Tables 6 to 11, the individual analysis results of the probands are listed.

Table 6. Analysis Results of Proband 1.

Proband 1	Reference Value	AGaMon Measurement	Relative Analysis Error (in %)
Meas. 1	0.29	0.24	20.2
Meas. 2	0.59	0.30	96.0
Meas. 3	0.66	0.35	90.2
Meas. 4	0.87	0.50	107.6
Meas. 5	0.91	0.54	110.9
Meas. 6	1.10	0.87	26.90

The mathematical calibration model was calculated with the calibration and evaluation program ProSens based on calibration measurements. This mathematical calibration model was underlaid for all measurements of the 6 probands. The evaluation was also performed with the program ProSens.

Table 7. Analysis Results of Proband 2.

Proband 2	Reference Value	AGaMon Measurement	Relative Analysis Error (in %)
Meas. 1	0.13	0.49	73.7
Meas. 2	0.32	0.27	18.3
Meas. 3	0.48	0.43	11.0
Meas. 4	0.64	0.56	13.6
Meas. 5	0.76	0.76	0
Meas. 6	-	-	-

Table 8. Analysis Results of Proband 3.

Proband 3	Reference Value	AGaMon Measurement	Relative Analysis Error (in %)
Meas. 1	0.28	0.31	10.3
Meas. 2	0.49	0.44	11.1
Meas. 3	0.83	0.49	67.8
Meas. 4	1.15	1.00	15.4
Meas. 5	1.32	0.53	147.8
Meas. 6	1.08	0.69	56.1

Table 9. Analysis Results of Proband 4.

Proband 4	Reference Value	AGaMon Measurement	Relative Analysis Error (in %)
Meas. 1	0.40	0.35	13.3
Meas. 2	0.66	0.57	15.5
Meas. 3	0.91	0.54	70.1
Meas. 4	1.11	1.04	6.4
Meas. 5	1.39	1.39	0
Meas. 6	1.23	1.33	7.6

Table 10. Analysis Results of Proband 5.

Proband 5	Reference Value	AGaMon Measurement	Relative Analysis Error (in %)
Meas. 1	0.37	0.33	11.2
Meas. 2	0.52	0.52	0
Meas. 3	0.61	0.62	1.0
Meas. 4	0.82	0.64	27.0
Meas. 5	1.05	1.16	9.5
Meas. 6	1.10	1.25	12.1

Table 11. Analysis Results of Proband 6.

Proband 6	Reference Value	AGaMon Measurement	Relative Analysis Error (in %)
Meas. 1	0.17	0.28	38.6
Meas. 2	0.28	0.37	23.6
Meas. 3	0.39	0.46	15.5
Meas. 4	0.57	0.57	0.4
Meas. 5	0.82	1.05	21.6
Meas. 6	0.84	1.33	36.7

The results of proband 1 in Table 6 shows that the AGaMon results are consequently to small compared to the results obtained by the reference measurements. The reason could be an erroneous handling of the proband or that the calibration model does not really fit the used AGaMon system. When not using the overall calibration model but individually calibrating the measurements of proband 1, there are very good analysis results.

The results of proband 2 in Table 7 shows very good analysis results. Only in measurement 1, there is a big discrepancy between AGaMon measurement and reference measurement. The reason could be an erroneous handling of the proband in the first measurement.

The results of proband 3 in Table 8 also shows rather good analysis results. The too small measurement results of measurements 3, 5 and 6 are certainly effected by erroneous handling of the proband.

The results of the probands 4 and 5 in the Table 9 and 10 show very good analysis results. The too small measurement results of measurement 3 of proband 4 and of measurement 4 of proband 5 are certainly effected by erroneous handling of the probands.

The results of proband 6 in Table 11 are not easy to explain because in all cases the measurement result obtained by the AGaMon system is much higher than the results obtained by the reference measurement. Maybe, this is due to the fact that the mathematical calibration model does not really fit the AGaMon system used by proband 6.

When not using the overall calibration model but individually calibrating the measurements of proband 6, there are very good analysis results.

Altogether, the results of the first field test show very promising analysis results. Of course, the sensor system AGaMon must be further enhanced and the reasons for the outliers must be investigated.

6. Conclusion and Future Work

An innovative mobile sensor system is developed, which is able to run a variety of commercially available metal oxide gas sensors in different parametriseable thermo-cyclic modes and can be exposed to several gases in the exhaled air. Therefore, this sensor system can be applied to several applications. In the application under consideration in this paper, the alcohol control in the exhaled air, ethanol is the leading component. But other interfering gases like H2 and acetone may occur in the air we exhale. Therefore, ternary ethanol-H_2-acetone mixtures have to be considered and analyzed. The sensor system, equipped with the metal oxide sensor MLV from Applied Sensors, operated in step-wise thermo-cyclic mode and with the incorporated advanced calibration and evaluation procedure ProSens 3.0, is an appropriated and powerful tool for this application. The analysis shows that very good substance identification can be achieved and the relative analysis errors of the concentration determination for the leading component ethanol is in all considered cases less than 9 %, even in the presence of interfering gases like H_2 and acetone.

The above obtained analysis results are based on measurements in the laboratory. First field tests, performed by 6 probands, show very promising results. In future work, the sensor system has to be further enhanced to meet the requirements of an applicable field sensor system.

Furthermore, the sensor system will be enhanced and adapted to further applications in the exhaled air like diabetes, asthma and halitosis. This would enable the sensor system to cover almost all significant aspects in human health care and medical applications.

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