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# New approaches for improving selectivity and sensitivity of resistive gas sensors: a review

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**Purpose** – The aim of the paper is to present methods of improving selectivity and sensitivity of resistance gas sensors.

**Design/methodology/approach** – We compare various methods of improving gas sensing by the temperature modulation, the UV irradiation or fluctuation enhanced sensing. We analyze low-frequency resistance fluctuations in commercial Taguchi gas sensors and the recently developed WO<sub>3</sub> gas sensing layers, exhibiting a photo-catalytic effect.

**Findings** – The efficiency of using low frequency fluctuations to improve the gas detection selectivity and sensitivity was confirmed by numerous experimental studies in commercial and prototype gas sensors.

**Research limitations/implications** – A more advanced measurement setup is required to record noise data but reduces the number of gas sensors necessary for identifying the investigated gas mixtures.

**Practical implications** – Fluctuation enhanced sensing can reduce the energy consumption of gas detection systems and assures better detection results.

**Originality/value** – A thorough comparison of various gas sensing methods in resistance gas sensors was presented and supported by exemplary practical applications.

**Keywords** Gas sensors, Noise, Low-frequency noise, Detection methods, Photo-catalytic effect

**Paper type** Research paper

## 1. Introduction

Resistive gas sensors—also known as Taguchi gas sensors—are very popular as a consequence of their reasonable price and good durability (Sberveglieri, 1995). However, their selectivity and sensitivity are limited, and these deficiencies serve as the main driving force to develop new materials and methods for the superior gas detection. Taguchi gas sensors comprise a porous semiconductor layer (e.g., SnO<sub>2</sub>, WO<sub>3</sub>, TiO<sub>2</sub>, ZnO) which starts to be gas sensitive at an elevated temperature, up to a few hundred °C (Fig. 1). This means that the sensors can be used to analyze even exhaust gases, but they have to consume energy for heating. Therefore, it is important to propose new methods which are able to improve the state-of-the-art in gas sensing.

There are numerous papers focused on optimizing parameters of metal oxides in order to enhance their selectivity and sensitivity (Li *et al.*, 1999; Franke *et al.*, 2006; Korotcenkov, 2005; Hyodo *et al.*, 2003; Yamazoe *et al.*, 1983; Heszler *et al.*, 2007). Thus, it was established that a vital improvement can be reached via controlling the size and the

shape of the gas sensor's oxide grains (Li *et al.*, 1999; Franke *et al.*, 2006) or by introducing catalytically active additives in the sensing layer via doping or a surface functionalization (Korotcenkov, 2005; Hyodo *et al.*, 2003; Yamazoe *et al.*, 1983; Heszler *et al.*, 2007). The results of these improvements are still far from satisfactory, and therefore it is of great interest to consider new methods for advancing the gas detection.

The gas detection efficiency can be enhanced not only by a technological development of the sensors but also by new measurement methods and signal processing. Recently, experimental studies confirmed that low-frequency, colored resistance fluctuations, generated within the gas sensors, can be utilized to improve their selectivity and sensitivity (Kish *et al.*, 2000; Vidybida, 2003; Ederth *et al.*, 2006; Kotarski *et al.*, 2009). It is well known that the  $1/f$  noise is intense in porous materials and depends on their surface treatment (Stadler *et al.*, 2014; Granqvist *et al.*, 2008). It is also clear that similar phenomena can be used to determine the ambient atmosphere for gas sensors. The low frequency noise can be characterized by various statistical functions (e.g. the power spectral density or the probability distribution); they are more informative than the DC resistance which gives nothing but an individual number for the sensor.

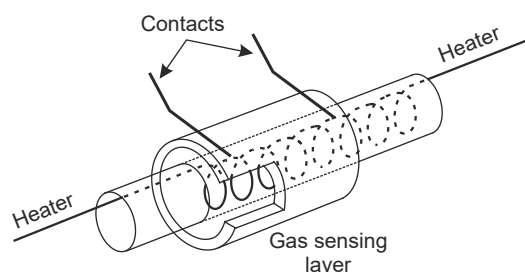


Fig. 1. The structure of a resistive gas sensor

Another method, which can be easily implemented to improve the gas detection, utilizes the temperature modulation of the gas sensing layer. Processes connected with the gas molecule adsorption and desorption depend on temperature and are responsible for a drop in the sensitivity at low and high operating temperatures (Morrison, 1987). The competition between adsorption and desorption processes determines the temperature of the maximum sensitivity and its decrease at temperature changes. This means that different temperature profiles should be observed for different gases and utilized for selectively recognizing them, as has been confirmed experimentally (Yamazoe *et al.*, 2011; Zaretskiy *et al.*, 2012). Moreover, a change of the gas sensitivity versus temperature ratio can be utilized to preserve the information about the ambient atmosphere within the gas sensor by its cooling in the investigated environment and “freezing” the gas molecules within the gas sensing layer for a further analysis under laboratory conditions at an elevated temperature. This technique is referred to as the “sample-and-hold” and can be used together with fluctuation enhanced sensing (Ayhan *et al.*, 2013).

Adsorption and desorption processes are accelerated when the gas sensing layer operates at an elevated temperature, typically above 150°C. However, such conditions limit possible sensor applications and furthermore increase the energy consumption. There is an experimental evidence that some gas sensing materials exhibit a photo-catalytic effect, and their sensitivity to various gases can be modulated by the UV irradiation (Comini *et al.*, 2000; Mishra *et al.*, 2004; Zaretskiy *et al.*, 2012; Chen *et al.*, 2012; Ayhan *et al.*, 2013). This effect has been observed, even at the room temperature, and resulted in a better sensitivity to the selected gases. We can expect that similar effects of the UV irradiation can be observed for the DC resistance and low-frequency noise as well. Preliminary experimental results confirmed the photo-catalytic effect for a very thin TiO<sub>2</sub> gas sensing layer, but its very high resistance limited practical applications (Topalian *et al.*, 2007). The resistance noise induced by the UV irradiation could potentially be a very sensitive tool, because the generated noise should be sensitive to the wavelength of the applied irradiation. Thus a combination of different wavelengths of the light sources could result in the superior gas selectivity as compared with other methods.

The mentioned sensing methods can provide an additional information about the ambient atmosphere of the gas sensor than what is possible with the DC resistance only. This information has to be properly processed to get satisfactory detection results by applying various algorithms. Usually, different gases are detected by applying a sensor array (electronic nose) and a processing array for DC resistances using pattern recognition methods (Machado *et al.*,

2005; Distant *et al.*, 2012; Kalinowski *et al.*, 2013). We will present a number of selected detection methods which can guarantee that the gathered noise data and dynamic changes of the DC resistance at a temperature change limit the number of sensors necessary to record the selected gases.

## 2. Fluctuation-enhanced gas sensing

Resistive gas sensors exhibit an intense  $1/f$  noise component, which overwhelms the white noise up to tens of kHz at least. The low-frequency noise is generated by fluctuations of the potential barrier between the grains (Fig. 2). The barrier depends on adsorption and desorption processes, and therefore its fluctuations can be selective to the ambient atmosphere of the gas sensor and its temperature. There is an experimental evidence that the  $1/f$  noise increases when the grains are smaller. Additionally, noise changes are much more sensitive than changes in the DC resistance, and the sensitivity increases further when the grains of the investigated gas sensing layer decrease (Ederth *et al.*, 2006).

The low-frequency noise generated within the porous gas sensing layer can be observed as voltage or current fluctuations across/through the gas sensor biased by a constant voltage  $U$  or current  $I$  in accordance with the Hooge formula (Hooge *et al.*, 1981):

$$\frac{S_u(f)}{U^2} = \frac{S_i(f)}{I^2} = \frac{\alpha}{fnV_{eff}}, \tag{1}$$

where  $S_u(f)$  and  $S_i(f)$  are power spectral densities of voltage and current fluctuations observed across/through the biased gas sensor, respectively,  $\alpha$  is the Hooge parameter which depends on the material of the gas sensing layer and the ambient atmosphere,  $n$  is the density of charge carriers involved in the noise generation within the gas sensing layer, and  $V_{eff}$  is the effective noise volume of the sensor determined by the grain boundary contacts. Thus, to get an information about the ambient atmosphere we have to estimate  $S_u(f)/U^2$  or  $S_i(f)/I^2$ , which are independent of the sensor bias. Figure 3 shows examples of noise data observed in a commercial TGS 826 gas sensor. Typically, we observe the  $1/f$  noise whose intensity depends on the ambient atmosphere. The power spectral density can change by as much as a few orders of magnitude and is characteristic for the investigated gas and its concentration.

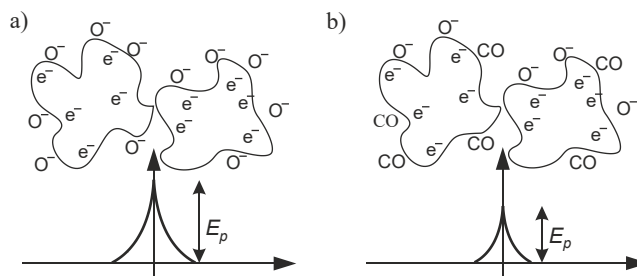


Fig. 2. The influence of the ambient atmosphere causing changes in the potential barrier  $E_p$  between the grains of the gas sensing layer: (a) in the presence of oxygen ions ( $O^-$ ) and (b) displaced by carbon monoxide (CO) molecules

The observed  $1/f$  noise can give a sufficient information to limit the number of sensors necessary to distinguish between the selected gases (e.g., ammonia and hydrogen sulfide), as presented by Kotarski and Smulko (2010). When a single gas sensor is applied, and one observes its low-frequency noise and DC resistance by measuring the DC voltage across the sensor (Fig. 4), it is possible to determine the gas concentration and detect gaseous species.

The power spectral density  $S_u(f)$  observed at the output of the system depends on the ambient gas by a change of the normalized spectrum  $S_u(f)/U^2$ , and — independently — on the change of the sensor's DC resistance  $R_s$  and the driving resistance  $R_1$  according to

$$S_u(f) = \frac{S_u(f)}{U^2} U^2 = \frac{S_u(f)}{U^2} U_0^2 \left( \frac{R_s}{R_1 + R_s} \right)^2. \tag{2}$$

When the sensor exhibits various changes of  $S_u(f)/U^2$  in different gases, one can establish the type of gas and the DC resistance determines its concentration. Equation (2) can be used for detection only because it depends on the measurement setup (resistance  $R_1$ ). The normalized power spectrum of the sensor resistance fluctuations  $S_{R_s}(f)/R_s^2$  is independent from the measurement system and can be estimated from the product  $S_u(f)/U^2$  as presented by Chang *et al.*, 2009. Figure 5 presents data observed for two gases, ammonia and hydrogen sulfide, in a commercial gas sensor TGS 826 (Kotarski *et al.*, 2010). The presence of hydrogen sulfide increases the power spectral density, whereas ammonia decreases it. Thus, using a single sensor one can detect which gas is present by estimating  $S_u(f)$  within the selected bandwidth and determining its changes by comparing them with measurements in the air.

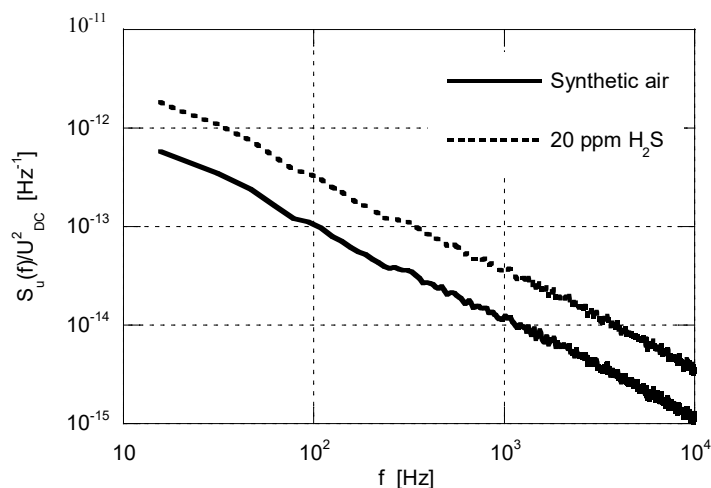


Fig. 3. The power spectral density  $S_u(f)$  of voltage fluctuations across a commercial TGS 826 gas sensor, normalized to the square of the DC bias voltage  $U$ . The data were taken in the ambient atmosphere of hydrogen sulfide ( $H_2S$ ) diluted in a synthetic air at different concentrations. The noise is observed within a few decades of the frequency  $f$

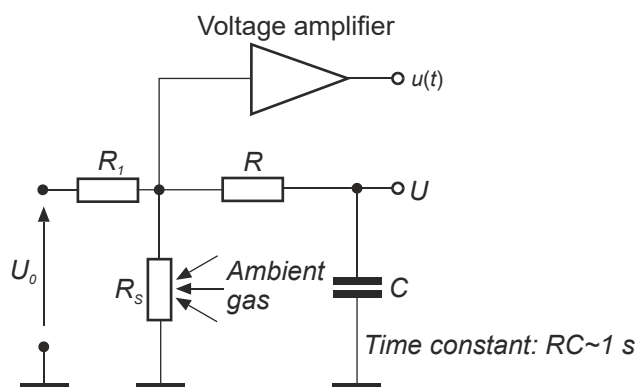


Fig. 4. The measurement set-up for the power spectral density  $S_u(f)$  of voltage fluctuations  $u(t)$  across a gas sensor having a DC resistance  $R_s$  and being biased by a DC voltage  $U$

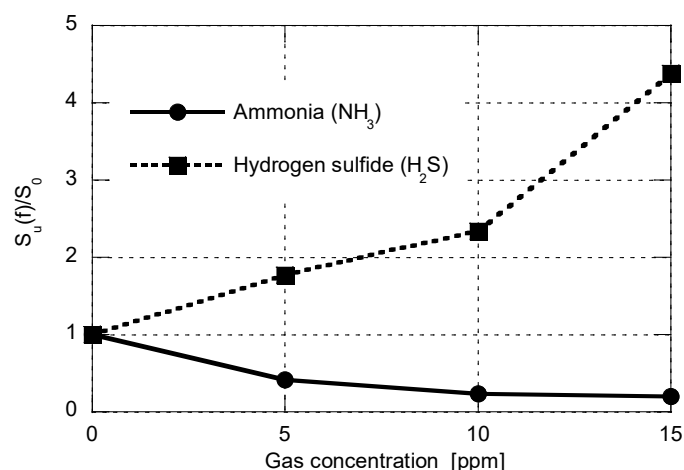


Fig. 5. The normalized power spectral density  $S_u(f)$  of voltage fluctuations across the sensor at 1kHz for a commercial TGS 826 sensor in various gases and concentrations. The sensor was serially connected with a resistor  $R_l = 11 \text{ k}\Omega$  and biased by a stabilized voltage source  $U_0$  (cf. Fig. 4)

Other methods for the gas detection by using fluctuation enhanced gas sensing require more advanced statistical parameters (e.g. bispectrum) in order to expose a non-Gaussian component, characteristic for various gases (Smulko *et al.*, 2001). A bispectrum function can measure the intensity of that component but requires more data to be estimated than those needed for the power spectral density. Cross-level images of the bispectrum can be used to determine the presence of different gases (Schmera *et al.*, 2010). There is an evidence that the non-Gaussian component is more intense with gas sensors comprising smaller grains (Smulko *et al.*, 2004). The bispectrum function is often much more informative than the power spectral density only, but the requirements of a longer averaging time and slower noise sampling limit its practical application.

### 3. Gas sensor temperature modulation

Features of resistive gas sensors depend strongly on their working temperature, and this fact can be used to gather more information about the ambient atmosphere in order to improve the sensitivity and selectivity. Typically, one can use pulse heating to clean the gas sensing layer by removing adsorbed molecules. When the intensity of the heating pulse is carefully selected, one can observe dynamic changes of the DC resistance  $R_S$  with a characteristic shape determined by the composition of the ambient atmosphere (Fig. 6). It should be underlined that a drop of the heating voltage from 5 to 4.7 V results in a significant change of the DC resistance but almost no change of  $S_u(f)/U^2$ . Thus, in order to improve the gas detection one should consider the shape of the DC resistance changes, or selected parameters describing them, such as the maximum and the derivative at a rise and a drop. Results analogous to those in Fig. 6 were observed when the heating voltage was increased; then the DC resistance dropped and exhibited a local minimum mirroring the shape of the time dependence shown in Fig. 6. Time constants of the DC resistance modulation depend on the gas sensors, their ambient atmosphere, and the temperature alteration. Thus, in some prototype gas sensors with very thin gas sensing layers, one can observe a much stronger change of the DC resistance and the absence of a peak structure (Fig. 7), which is a consequence of a smaller thermal inertia than that for a commercial sensor (Fig. 6) with a much greater volume (Fig. 1).



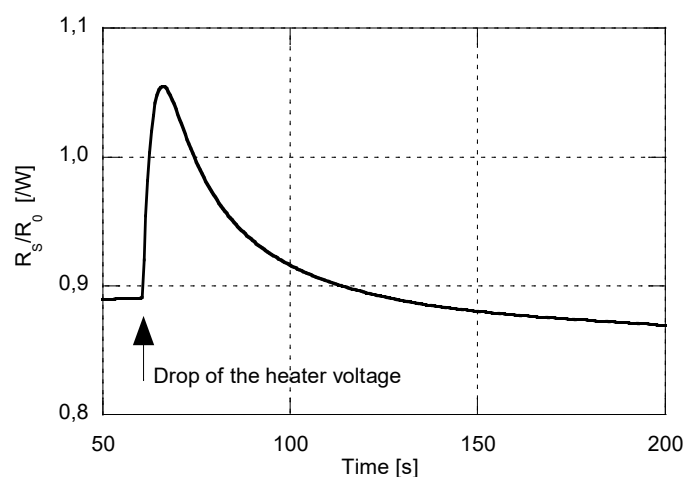


Fig. 6. The time-dependent gas sensor DC resistance  $R_S$  at an abrupt drop of the heating voltage from 5 to 4.7 V. A commercial gas sensor (TGS 2600) was placed in the ambient atmosphere of combustible gases ( $C_2H_6$ ,  $C_3H_8$ ,  $n-C_4H_{10}$ ,  $C_4H_{10}$ );  $R_S$  was normalized to the sensor's DC resistance  $R_0$  observed in a synthetic air

It is worth mentioning that the temperature modulation can lead to very interesting and new applications when the heated gas sensor is exposed to an unknown gas mixture and subsequently cooled. The gas molecules will then be trapped inside the porous gas sensing layer as a result of less intense adsorption/desorption processes and preserved for a further analysis under laboratory conditions (Ayhan *et al.*, 2013). Moreover, one can expect that the resistance noise in the gas sensing structure could be observed and used for the gas detection even at temperatures lower than those typically employed for the gas sensor. This means that various temperatures could be applied to establish an adsorption–desorption profile of the explored atmosphere and providing an improved gas detection.

#### 4. Light-induced gas sensing

The light activation for gas sensing applications has been reported for various oxide layers (e.g., ZnO, WO<sub>3</sub>, SnO<sub>2</sub>, TiO<sub>2</sub>) in literature (Ayhan *et al.*, 2000; Mishra *et al.*, 2004; Topalian *et al.*, 2007; Chen *et al.*, 2012). The UV light is applied to assure a sufficient energy for acceleration of adsorption/desorption processes. Usually the same effect can be obtained by heating the gas sensing layer. This means that using the UV light results in a lower energy consumption and ability to work at much lower temperatures. The second fact opens new applications for resistive gas sensors (e.g. detecting combustible gases or explosives). Additionally, one can expect that the UV light reduces the measurement time and can be used to clean gas sensors after poisoning (Comini *et al.*, 2000).

Examples of data recorded with an Au-nanoparticle-decorated WO<sub>3</sub>-nanowire gas sensing layer are presented in Fig. 7 (Vallejos *et al.*, 2013), which compares the change of the sensor's DC resistance, induced by an abrupt increase of the heating voltage, with that caused by the UV irradiation using a T5F UV diode emitting radiation at a wavelength of 365 nm. The diode was placed about 5 mm from the surface of the sensor layer, and its DC current  $I_d$  was controlled up to the maximum value of 20 mA.

One can conclude that additional data on the ambient atmosphere can result from the application of an abrupt temperature change as well as from the UV irradiation. The shape of the recorded curve depends on the sensor's ambient atmosphere, as was observed for a synthetic air (Fig. 7) and ethanol (Fig. 8). Data collected by using the UV light and temperature modulation are rather similar, at least for the presented results, but it should be underscored that the UV light yields a lower energy consumption than that necessary for the temperature modulation (the effect on the sensor's DC resistance change can be observed even at low diode currents  $I_d$ ; *cf.* Fig. 8). Additionally, using the UV light one can detect gases at much lower temperatures, which is a very attractive feature. One can expect that the UV irradiation will increase the usefulness of fluctuation enhanced sensing, because the  $1/f$  noise might depend strongly on the wavelength

of the applied UV light and therefore should provide more information about the ambient gas than the temperature modulation alone.

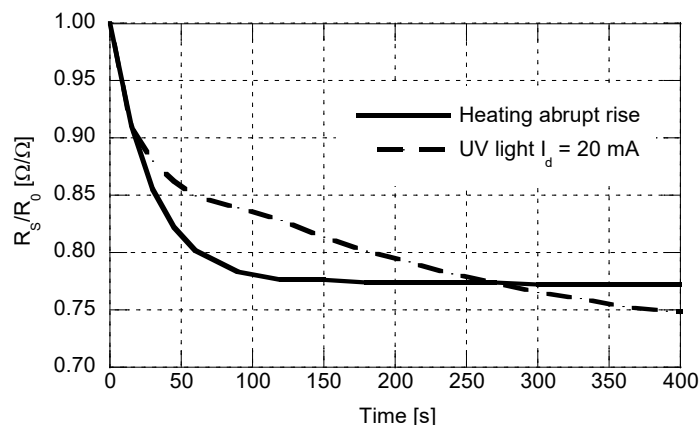


Fig. 7. The relative change of the DC resistance  $R_S$  for a  $\text{WO}_3$  gas sensing layer at an abrupt rise of the heating voltage from 1.5 to 1.8 V, corresponding to the temperature change between 100 and 120°C (solid curve), and by the UV irradiation (dashed curve).  $R_0$  denotes the resistance in a synthetic air and at the heating voltage of 1.5 V, and  $I_d$  is the UV diode current

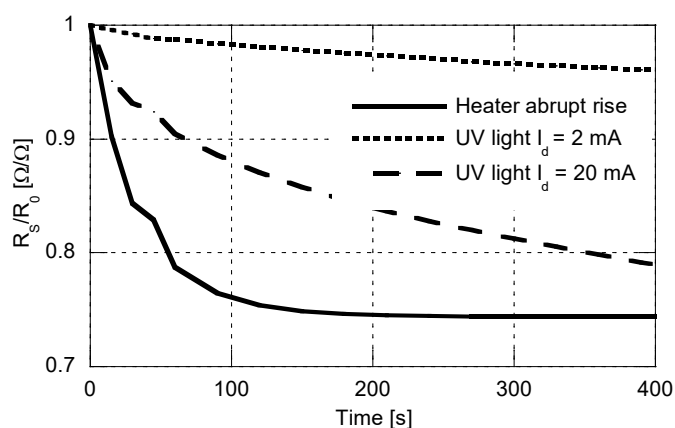


Fig. 8. The relative change of the DC resistance  $R_S$  for a  $\text{WO}_3$  gas sensing layer at an abrupt rise of the heating voltage from 1.5 to 1.8 V, corresponding to the temperature change between 100 and 120°C (solid line), and by the UV irradiation at the shown values of the UV LED current. Data were taken in 200 ppm of ethanol.  $R_0$  denotes the resistance in 200 ppm of ethanol at the heating voltage of 1.5 V

## 5. Detection algorithms

This paper has demonstrated that gas sensing can in some cases be improved by very simple methods, and different gases can be detected by a single gas sensor by using noise measurements (*cf.* Fig. 5). In general, the same detection algorithms can be used for gas sensing as for other detection methods, e.g. chemometric techniques as presented by Geladi *et al.*, (2005) when the input data are vectors. A set of DC resistances, the power spectral density of voltage fluctuations across the sensor (Fig. 3), and a dynamic change of the DC resistance in selected time intervals (Fig. 6) can be treated as vectors. Such data can be analyzed by a principal component analysis (PCA), which utilizes the linear and orthogonal data transformation into a new set of linearly uncorrelated variables, referred to as “principal components”. This method requires algebraic computations to perform the necessary transforms in order to establish the most informative components. The method is commonly used in gas sensing applications for the gas detection or for the gas

concentration prediction. A more advanced method employs a support vector machine (SVM) algorithm, which is a nonlinear version of the PCA (Distante *et al.*, 2003; Kalinowski *et al.*, 2013). The latter method gives even better results, because gas sensors respond to various gases in a nonlinear manner. Both PCA and SVM can be applied to detect the presence of the selected gases which exceed threshold concentrations and to establish their concentration using a regression model.

The data used for the gas detection (e.g. the power spectral density) are vectors whose adjoining elements are correlated, and therefore methods such as PCA or SVM have to reduce abundant data. Therefore, other methods which might have similar detection efficiency but require much less intense computations, can be of interest. Such methods use selected parameters of the estimated functions. In the case of the power spectral density, one can use its intensity and slope within a selected bandwidth (Chang *et al.*, 2009). This method requires a limited computation and can create a reduced set of data for further processing by a more computationally demanding method, e.g. an SVM algorithm (Turner and Magan, 2004).

In general, the algorithms have to be adjusted for the case when a few volatile components have to be detected and additionally some specific substances exist in the ambient atmosphere and conceal the effects caused by the chemicals of interest. A good example is the identification of chemical compounds in a human breath, which can be characteristic for some diseases (Machado *et al.*, 2005).

## 6. Conclusions

Various methods for improving the selectivity and sensitivity of resistive gas sensors were presented. The proposed methods can be applied to prepare more advanced measurement set-ups than those commonly used today. The same techniques can also be used to improve new gas sensing materials. A combined method of fluctuation enhanced sensing and the UV light modulation might be the most promising method to reach the maximum sensitivity and selectivity at a limited cost, as also supposed by other authors (Lay-Ekuakille *et al.*, 2014).

Table 1. Pros and cons of the considered gas sensing methods

Method	Advantages	Disadvantages
Resistance	Easy measurements; can be modulated by temperature change or UV light when photo-catalysis is present.	Gives a single number only; requires additional sensors to detect composition of gas mixtures.
Fluctuation enhanced sensing	Up to a few hundred times more sensitive than DC resistance; gives reduction of consumed energy.	More complicated measurements and estimation method; longer averaging time.

It is difficult to compare the efficiency of fluctuation enhanced sensing with the gas detection using DC resistance measurements only because both methods establish a different information about the sensor's ambient atmosphere. The main features for comparing both methods are collected in Table 1. We can conclude in general that fluctuation enhanced sensing is more sensitive and gives an additional information when compared with DC resistance measurements only. Additionally, differences between the methods are more visible when the sensors are comprised of very small grains. Then the gas sensors respond to gas changes in a faster way, and the  $1/f$  noise predominates even up to tens of kHz (Ederth *et al.*, 2006; Smulko *et al.*, 2004; Kwan *et al.*, 2008).

The presented new approaches to gas sensing using resistance sensors should facilitate the introduction of new applications by enabling the gas detection at different temperatures than previously assumed or determining the composition of the gas mixture with a reduced number of the applied sensors.

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