

THE DERIVATION OF THE ELECTRICAL CONDUCTANCE/TEMPERATURE DEPENDENCY FOR TIN DIOXIDE GAS SENSOR

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Abstract. *The dependency between the electrical conductivity and the temperature of the tin dioxide layer at the constant concentration of the detected gas is derived here. The derived equation is modified as a function of two variables describing the dependence of the electrical conductivity of the sensor on both the temperature and the concentration. The derived formulas were verified by approximation of the measured data. These formulas can be useful in practical applications.*

Keywords

Electrical conductance, gas sensor, modelling, tin dioxide.

1. Introduction

The tin dioxide sensors for the gas detection in air have been used for years. The detection process is based on the changes of the electrical conductivity of a tin dioxide layer heated to a sufficiently high temperature. Sensor response - it is a term used here to refer to the electrical conductance of the tin dioxide layer. The metal oxide detection layer is a polycrystalline structure. This is why the physical-chemical phenomena appearing on the surface of the layer are complex and it is very difficult to build the mathematical description of the behaviour of the sensor during the detection of the substance.

There are several methods for the description of the sensor behaviour. One group of the methods is based on the band theory. On its base the equations describing the electrical conductance of the sensor are derived in [1], [2], [3]. The advantage of the above-mentioned description is that it is general and universal. The disadvantage is its low objectivity and weak coherence

with physical-chemical phenomena during the detection. The second possibility is using a pure mathematical approximation of the measured data. It is shown for example in [4], [5], [6]. The advantage of this approach is that it is not necessary to deal with the complex physical-chemical phenomena or the band theory. But the obtained equations are without any physical relation to the object being described. The third group deals with the physical-chemical phenomena. The examples of such an approach can be found in [7], [8], [9], [10], [11], [12], [13], [14], [15], [16], [17]. The advantages of this description are the objectivity and the relation to the physical-chemical phenomena. So the sensor behaviour may be better understood from the macroscopic point of view. On the other hand, the complexity of the obtained formulas and their accordance with the experimental values depends on the choice of the phenomena used for the behaviour of the sensor at the given conditions. This article describes the sensor response in terms of physical-chemical phenomena under simplified assumptions. The aim of this work is to conceive a simple model of the sensor behaviour, that fits in the sensor response and that can be useful for the sensor applications. This model extends the results described in [18].

2. Experiments

The commercial tin dioxide sensors of type TGS 813, TGS 822, SP 11 and MQR 1003 were chosen for the experiments. The sensors TGS 813, SP 11 and MQR 1003 are meant for detection of methane. TGS 822 is meant for the detection of ethanol, acetone and organic solvents. Different technologies were used for production of the sensors, so they have different levels of selectivity and sensitivity. The vapour of acetone, benzene, ethanol and hexane were used in the experiments, always only one compound in air. Each substance was

tested by all sensors at the same time to ensure the equal concentration of the tested vapours and the equal value of the heating voltage for all sensors. The saturated vapour above the liquid was obtained at a constant ambient temperature of 25 °C. The desired concentration was prepared by injecting of a certain volume of saturated vapour into the tested chamber filled with the air of a known volume of 2700 ml according to the Antoine equation. The heating voltage was set to $U=5$ V and the sensors were inside the testing chamber in a flux of clean air before every measurement so as to remove the adsorbed undesired substances from the surface of SnO_2 [19], [20]. The used laboratory air was purged of dust and its relative humidity was decreased to RH=25 %. The recovery of the sensors was completed when the sensor response became practically independent of time. Related value of the sensor response never exceeded 10 μS . Afterwards, the heating voltage was decreased to $U = 2$ V by a step change to start sensing process, but to keep suitable temperature of the layer to avoid adsorption of water to the surface of SnO_2 . According to the previous experience, the value of $U = 2$ V corresponds to the surface temperature of approximately 500 K. After that the tested vapour was injected into the chamber and the measurement was carried out. The heating voltage was increased step by step by 20 mV up to 5 V. The electrical conductance was sensed 10 s after the change of the heating voltage. The time of 10 s was established experimentally as the moment at which the value of the conductivity is practically independent of time. The electrical circuit for sensing the conductivity of the sensor was used according to the data sheet recommendations of the producers of the sensors. The resistor of $R = 2$ k Ω in series with the sensor and the supply voltage of $U = 5$ V were used. The temperature of the detection layer relevant to the range of the heating voltage of 2 V up to 5 V was estimated according to the temperature dependency of the alloy NiCr electrical resistance [21], which is widely used for the heating system of the given sensors. Furthermore, this estimated temperature was compared to the data from [22], where the temperature range for the detection of ethanol is described. Considering this, the temperature range related to the heating voltage $U = 2$ V up to 5 V corresponded to $T = 500$ K up to $T = 700$ K. The measurements were carried out at different values of concentration of the tested substances in air. The temperature characteristics of the sensor - it is a term used here to refer to the dependency of the electrical conductivity of the sensor on the temperature at the constant concentration.

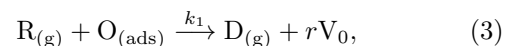
3. Theory

Chemisorption of gases on the surface of a tin dioxide detection layer can be considered as monomolecular adsorption. So the process of chemisorption can be described by the Langmuir isotherm. On the basis of this, the electrical conductivity of the sensor Eq. (1) was derived in [18].

$$G(x) = G_0 + Z \frac{Kx}{1 + Kx}, \quad \text{where } x = \frac{p_R}{p_T} 10^6, \quad (1)$$

$$K = b \frac{p_T}{10^6}, \quad Z = \frac{e N_A k_1 m_1 a_{\max}}{U}, \quad (2)$$

where x is the relative concentration of the detected gas in ppm, G_0 is the basic electrical conductivity of the sensor in clean air, that is at $x = 0$, b is the adsorption coefficient in Pa^{-1} , p_T is the total pressure of the mixture in Pa, e is the electron electrical charge in C, N_A is the Avogadro constant in mol^{-1} , k_1 is the reaction rate constant in s^{-1} , m_1 is the mass of the detection layer SnO_2 in kg, a_{\max} is the adsorption capacity of SnO_2 in $\text{mol} \cdot \text{kg}^{-1}$, U is the used measuring voltage in V and it is always kept constant during the experiments. It is assumed in Eq. (1) that the temperature of the detection layer is constant while the concentration x is a variable. In this article this equation is referred to as the transfer characteristics of the sensor. The examples of the reaction mechanisms of some substances on the surface during the detection are described in [23], [24], [25], [26], [27], [28]. Equation (1) is derived under the assumption that a system of subsequent reactions on the surface can be commonly expressed by one symbolic equation



where R represents the reducing gas being detected, O the adsorbed oxygen on the layer of SnO_2 , D represents the final products formed during the detection, V_0 are the oxygen vacancies (the released electrons changing the electrical conductivity of the sensor), r is a rational number.

The derivation described herein is based on the hypothesis, that the system of subsequent reactions during the detection can be represented by one symbolic reaction and the reactions during the detection in mentioned range of temperatures release free electron. This electron increases the electrical conductivity of the tin dioxide. Next hypothesis is, that the adsorption process of the detected substance and the desorption process of the reaction product are the features both for the transfer characteristic and for the temperature characteristic. All hypotheses and assumptions are supported with the experimental results presented herein.

Since there is a sufficient amount of the detected substance contained in the air surrounding of the sensor, the electrical conductivity of SnO₂ reaches the value that is relevant to the dynamic equilibrium at the given conditions. This value is practically independent of time.

It is necessary to modify Eq. (1) to be used for the derivation of temperature characteristics of the sensor. The concentration x will be constant and the temperature T of the detection layer of SnO₂ will be a variable. It means that coefficients G_0 , Y and K become dependent on the temperature T . In general, the temperature dependency of the conductivity of a detection layer exists in clean air $x = 0$ as well [1]. The temperature dependencies of the conductivities of the tested sensors were found experimentally and they do not exceed the values of 10 μS at $x = 0$ in the range of $T = 500$ K up to 700 K. That value can be neglected compared to the usual values of the sensor response (100 μS up to 1000 μS) for used range of concentrations and range of temperatures. So the term G_0 in formula Eq. (1) can be neglected.

The temperature dependency of coefficients Z and K in Eq. (1) can be derived by using the kinetic theory of gases. Relative coverage of a surface is used for gas adsorption on a solid surface and it is defined by

$$\Theta = \frac{N}{N_t} = \frac{a}{a_{\max}}, \tag{4}$$

where N is the number of species adsorbed on the surface, N_t is the total number of the adsorption sites on the surface, a is the adsorption coefficient mol · kg⁻¹, a_{\max} is the adsorption capacity of the surface mol · kg⁻¹. The adsorption process of the substance on the surface can be described by

$$\frac{d\Theta}{dt} = SF\sigma_0, \tag{5}$$

where S is the sticking coefficient, F is the number of the molecules striking the surface equal one per the time equal one, σ_0 is the area of one adsorption site. The Hertz Knudsen equation is valid for the ideal gas:

$$F = \frac{p}{\sqrt{2\pi m_1 k T}}, \tag{6}$$

where p is the gas pressure, m_1 is the mass of one gas molecule, k is the Boltzmann constant, T is the thermodynamic temperature. The sticking coefficient represents the probability that the struck molecule will remain adsorbed on the surface and will not get back to the gas phase. The coefficient is dependent on the temperature and on the coverage of the surface. In case that one molecule occupies only one adsorption site the formula is as follows:

$$S = S_0(1 - \Theta)e^{-\frac{E_A}{RT}}, \tag{7}$$

where S_0 is the probability of occupation of one site in case of a clean, still uncovered surface, that is $\Theta = 0$. S_0 usually equals 1. E_A is the activation energy of adsorption of gas in J · mol⁻¹, R is the universal gas constant, T is the thermodynamic temperature. For desorption of the substance out of the surface of coverage Θ , the Wigner Polanyi equation is used having the following form for the reaction of the first order:

$$\frac{d\Theta}{dt} = -k_2\Theta \quad \text{where} \quad k_2 = \nu_0 e^{-\frac{E_d}{RT}}, \tag{8}$$

where E_d is the activation energy of desorption of the adsorbed substance in J · mol⁻¹, R is the universal gas constant, T is the thermodynamic temperature, k_2 is the rate constant of the reaction. ν_0 is the frequency factor in the Arrhenius equation. For the frequency factor the approximative formula according to the Frenkel equation [29] is valid:

$$\nu_0 \doteq \frac{kT}{h} \quad \frac{1}{\nu_0} = \tau_0, \tag{9}$$

where k is the Boltzmann constant, h is the Planck constant, T is the thermodynamic temperature, ν_0 represents the frequency of vibration of one molecule chemisorbed on the surface. If the energy delivered to amount-of-substance of 1 mol equals to or is greater than E_d , the bond between the surface and the molecule is broken and the molecule desorbs to the gas phase. Time τ_0 in [30] is referred to the time for which the molecule remains on the surface before leaving it. For the equilibrium state between adsorption and desorption according to Eq. (5) and Eq. (8) there is

$$SF\sigma_0 - k_2\Theta = 0. \tag{10}$$

By substitution from Eq. (6), Eq. (7) and Eq. (9) to Eq. (10) the following equation is obtained:

$$\frac{p(1 - \Theta)\sigma_0}{\sqrt{2\pi k m_1 T}} e^{-\frac{E_A}{RT}} = \frac{1}{\tau_0} \Theta e^{-\frac{E_d}{RT}}. \tag{11}$$

The equation can be modified into the form of

$$K_A(1 - \Theta)p = K_D\Theta \tag{12}$$

$$K_A = \frac{\sigma_0 e^{-\frac{E_A}{RT}}}{\sqrt{2\pi m k T}} \quad K_D = \frac{1}{\tau_0} e^{-\frac{E_d}{RT}}. \tag{13}$$

From formula Eq. (12) the coverage of the surface Θ can be calculated:

$$\Theta = \frac{\frac{K_A}{K_D} p}{1 + \frac{K_A}{K_D} p} = \frac{bp}{1 + bp}. \tag{14}$$

Formula Eq. (14) is the Langmuir isotherm. Coefficient b can be calculated by substitution of K_A and K_D from Eq. (13) and we get

$$b = \frac{K_A}{K_D} = \frac{\tau_0 \sigma_0}{\sqrt{2\pi m_1 k T}} e^{\frac{E_d - E_A}{RT}} = \frac{\tau_0 \sigma_0}{\sqrt{2\pi m_1 k T}} e^{\frac{\Delta E}{RT}}, \tag{15}$$

by using the following formulas

$$k = \frac{R}{N_A} \quad m_1 = \frac{M}{N_A}, \quad (16)$$

formula Eq. (15) can be modified to the form mentioned in [30], or in [31], that is to

$$b = \frac{\tau_0 \sigma_0 N_A}{\sqrt{2\pi MRT}} e^{\frac{\Delta E}{RT}}, \quad (17)$$

where ΔE is the heat necessary for desorption of 1 mol of amount-of-mass of the substance (it practically equals the negative value of the adsorption heat), M is the molar mass of the adsorbed substance, N_A is the Avogadro constant, σ_0 is the area of one adsorption site, τ_0 is the time for the duration of which the adsorbed molecule stays on the surface, R is the universal gas constant, T is the thermodynamic temperature. At this moment the temperature dependency of coefficients Z and K in Eq. (2) can be derived. The Arrhenius equation is valid for the temperature dependency of the rate constant

$$k_1 = \nu_0 e^{-\frac{E_1}{RT}} \quad \text{where} \quad \nu_0 = \frac{kT}{h}, \quad (18)$$

where E_1 in $\text{J} \cdot \text{mol}^{-1}$ is the activation energy of the chemical reaction in Eq. (3). The following equation will be valid for Z after substitution of Eq. (18) into formula Eq. (2):

$$Z = \frac{e N_A k T m_1 a_{\max}}{h U} e^{-\frac{E_1}{RT}}. \quad (19)$$

After substitution b from Eq. (17) and τ_0 from Eq. (9) and their inserting into Eq. (2) we obtain the following equation for K :

$$K = \frac{p_T h \sigma_0 N_A}{10^6 k T \sqrt{2\pi MRT}} e^{-\frac{\Delta E}{RT}}. \quad (20)$$

And finally, after substitution Eq. (19) and Eq. (20) into Eq. (1), the equation for the temperature dependency of the detection layer will be in the following form:

$$G(T) \doteq Z \frac{Kx}{1 + Kx} = C_0 T e^{\frac{C_1}{T}} \frac{C_2}{T\sqrt{T}} e^{\frac{C_3}{T}}, \quad (21)$$

where for coefficients C_0 , C_1 , C_2 and C_3 we get

$$C_0 = \frac{e N_A k m_1 a_{\max}}{h U} \quad C_1 = \frac{-E_1}{R}, \quad (22)$$

$$C_2 = \frac{\sigma_0 N_A h p_T x}{10^6 k \sqrt{2\pi MR}} \quad C_3 = \frac{\Delta E}{R}. \quad (23)$$

Thus, from formulas Eq. (19) and Eq. (22) the following formulas emerge for coefficients Z and C_0 :

$$\gamma_1 = \frac{Z}{C_0} = T e^{\frac{-E_1}{RT}} = T e^{\frac{C_1}{T}}, \quad (24)$$

and from formulas Eq. (20) and Eq. (23) the following formulas are valid for coefficients K and C_2 :

$$\gamma_2 = \frac{K}{C_2} = \frac{1}{x T \sqrt{T}} e^{\frac{\Delta E}{RT}} = \frac{1}{x T \sqrt{T}} e^{\frac{C_3}{T}}. \quad (25)$$

4. Results

Equation (21) was used for approximation of the measured values of the electrical conductance of a tin dioxide layer at the concentration level $x = 600$ ppm at the temperature from 500 K up to 700 K. The computer programme XYMATH was used for the approximation by means of the least square method and the total error option was chosen as well. The correlation coefficient varied around $r = 0.98$ and the standard deviation was approximately 8 percent. That is acceptable. The example of approximation of the measured data for hexane and the used sensors are in Fig. 1, the example for different substances and sensor TGS 813 are in Fig. 2. The approximation of the measured data for different concentration values of acetone and sensor TGS 813 are in Fig. 3. These examples demonstrate good approximation ability of measured data by Eq. (21).

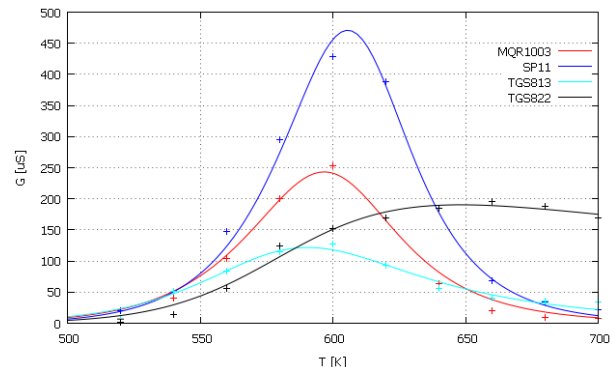


Fig. 1: The temperature dependencies of electrical conductivity G of the tested sensors for the concentration $x = 600$ ppm of hexane. The curves are approximations, the indicated points are the measured values.

The numerical values of the approximation coefficients of the temperature characteristics of the tested substances are in Tab. 1.

It is apparent from Tab. 1 that coefficients C_0 up to C_3 differ from each other. Coefficient C_0 is the highest and coefficient C_2 is the smallest one. The values of C_1 and C_3 are ordinarily comparable and the sign is opposite. C_1 is related to the activation energy of reaction Eq. (3) of the detected gas and oxygen. This value is always negative as, according to the Arrhenius equation, the temperature increases its value the reaction rate increases. The value of C_3 is related to the adsorption heat of the substance adsorbed on the surface of SnO_2 . The value of the adsorption heat is negative because chemisorption is an exothermic process. It is

Tab. 1: The coefficients of approximations of the temperature dependency of the electrical conductance of the sensors for $x = 600$ ppm.

	C_0 ($\mu\text{S} \cdot \text{K}^{-1}$)	C_1 (K)	C_2 ($\text{K}^{3/2}$)	C_3 (K)
MQR 1003				
acetone	$3.07 \cdot 10^{10}$	$-1.36 \cdot 10^4$	$4.11 \cdot 10^{-18}$	$2.92 \cdot 10^4$
benzene	$1.05 \cdot 10^{13}$	$-1.78 \cdot 10^4$	$3.79 \cdot 10^{-25}$	$3.94 \cdot 10^4$
ethanol	$1.30 \cdot 10^9$	$-1.13 \cdot 10^4$	$1.43 \cdot 10^{-11}$	$1.92 \cdot 10^4$
hexane	$9.85 \cdot 10^7$	$-1.12 \cdot 10^4$	$1.02 \cdot 10^{-16}$	$2.79 \cdot 10^4$
SP 11				
acetone	$1.17 \cdot 10^{10}$	$-1.28 \cdot 10^4$	$4.06 \cdot 10^{-22}$	$3.52 \cdot 10^4$
benzene	$1.58 \cdot 10^8$	$-1.10 \cdot 10^4$	$2.47 \cdot 10^{-18}$	$3.00 \cdot 10^4$
ethanol	$1.29 \cdot 10^6$	$-7.61 \cdot 10^3$	$1.48 \cdot 10^{-14}$	$2.42 \cdot 10^3$
hexane	$3.22 \cdot 10^9$	$-1.31 \cdot 10^4$	$2.97 \cdot 10^{-20}$	$3.32 \cdot 10^4$
TGS 813				
acetone	$1.34 \cdot 10^8$	$-1.06 \cdot 10^4$	$3.35 \cdot 10^{-16}$	$2.70 \cdot 10^4$
benzene	$1.37 \cdot 10^6$	$-8.76 \cdot 10^3$	$4.64 \cdot 10^{-13}$	$2.30 \cdot 10^4$
ethanol	$5.03 \cdot 10^4$	$-5.94 \cdot 10^3$	$4.70 \cdot 10^{-12}$	$2.10 \cdot 10^4$
hexane	$1.09 \cdot 10^7$	$-1.00 \cdot 10^4$	$3.34 \cdot 10^{-10}$	$1.84 \cdot 10^4$
TGS 822				
acetone	$9.33 \cdot 10^5$	$-1.72 \cdot 10^3$	$1.88 \cdot 10^{-8}$	$1.70 \cdot 10^4$
benzene	$3.56 \cdot 10^{11}$	$-1.64 \cdot 10^4$	$1.92 \cdot 10^{-9}$	$1.78 \cdot 10^4$
ethanol	$1.71 \cdot 10^9$	$-1.14 \cdot 10^4$	$4.22 \cdot 10^{-9}$	$1.63 \cdot 10^4$
hexane	$8.41 \cdot 10^8$	$-1.26 \cdot 10^4$	$1.12 \cdot 10^{-6}$	$1.34 \cdot 10^4$

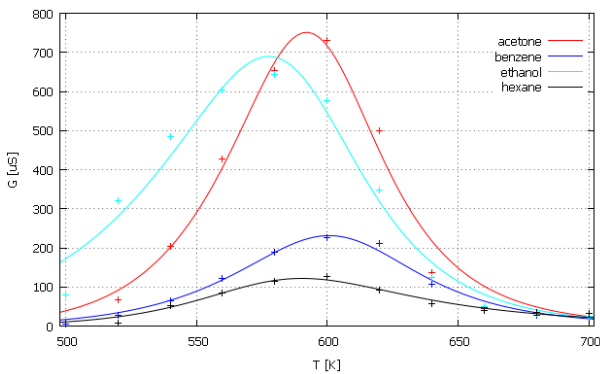


Fig. 2: The temperature dependencies of the electrical conductivity G of sensor TGS 813 for the concentration $x = 600$ ppm for different substances. The curves are approximations, the indicated points are the measured values.

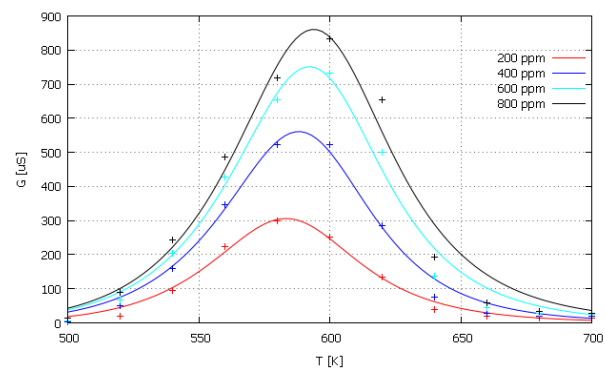


Fig. 3: The temperature dependencies of electrical conductivity G of sensor TGS 813 for different concentrations of acetone in air. The curves are approximations, the indicated points are the measured values.

necessary to supply this heat to the chemisorbed substance during the process of desorption to enable the substance to leave the surface. That is why coefficient C_3 is positive. Coefficients C_0 are ordinarily of high values and, on the other hand, coefficients C_2 are ordinarily of very low values. It is related to the values of the constants that occur in formulas Eq. (22) and Eq. (23). It emerges from Tab. 1 that the values of the coefficients depend both on the type of a sensor and on the type of a detected substance. The values of coefficients C_0 up to C_3 form a set of four values. This set is typical for the given substance, sensor and concentration. The set of four coefficients can help to discriminate the detected substance.

On the base of the derived formula Eq. (21) and its verification by the experimental results it is possible to suggest simplified behaviour of the sensor during the detection. If temperature of the layer increases its value the rate constant k_1 of reaction Eq. (3) increases. That is why the number of the released electrons increases. This leads to increasing the conductivity of the layer of SnO_2 . When the maximum value of conductivity is reached, the sensor response starts to decrease together with the rise of the temperature. Behind the maximum value of the sensor response the desorption influence begins to prevail since the rate constant k_2 in Eq. (8) increases its value together with the rising temperature. The molecules of the adsorbed substance oscillate more intensively and it is more difficult for them

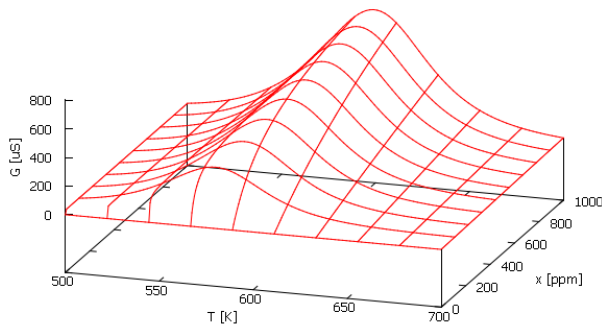


Fig. 4: The dependency of electrical conductance G of sensor TGS 813 on temperature T and gas concentration x for acetone.

to remain on the surface. Their number decreases with the rising temperature and due to this the amount-of-substance R in Eq. (3) decreases. That is why the number of the electrons released by chemical reaction Eq. (3) decreases as well and the conductivity of the layer decreases. These phenomena form the bell shape of the temperature characteristics, which is typical for most of the detected substances. Some substances have the increasing dependency only at the tested range of the temperature as apparent for hexane in Fig. 1. The hypothesis for explanation is, that the maximum of the detection curve of these substances is shifted towards the higher temperature that lies out of the tested temperature range. The maximum value of the response theoretically exists according the Eq. (21), but the observation of this maximum is limited by the maximum permissible value of the heating voltage so as not to damage the sensor.

Equation (21) manifests the typical features of the sensor. Then one equation of two variables can be formed from formulas Eq. (1) and Eq. (21):

$$G(T, x) = C_0 T e^{\frac{C_1}{T}} \frac{\frac{C_2^*}{T\sqrt{T}} x e^{\frac{C_3}{T}}}{1 + \frac{C_2^*}{T\sqrt{T}} x e^{\frac{C_3}{T}}}$$

where $C_2^* = \frac{C_2}{x_0}$, (26)

where x_0 is the reference value of the concentration. The dependency of the electrical conductivity of sensor TGS 813 for ethanol is in Fig. 4. The coefficients from Tab. 1 and the reference value $x_0 = 600$ ppm were used here.

5. Conclusion

The equation describing the temperature dependency of the electrical conductance of a tin dioxide layer for

the detection of the reducing gas is derived here. The derivation is based on the physical-chemical phenomena. Next research can be focused to theoretical justification of the simplified assumptions used herein. The derived equation was successfully verified by comparing with the experimental data. Reached results can be useful in practical applications.

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