ISSN 2616-8685. JOURNAL OF PHYSICS AND ELECTRONICS Vol. 32(1-2), 2024. DOI 10.15421/332409

ANALYSIS OF KINETIC DEPENDENCIES OF THE RESPONSE OF A GAS SENSOR BASED ON A MODEL OF A STRETCHED EXPONENTIAL FUNCTION WITH THE USE OF COMPUTER TECHNOLOGIES

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The results of improving the software of an information measuring system for studying relaxation phenomena in various objects based on a model of a stretched exponential function by supplementing it with intelligent components that eliminate operator participation are present.

A variant of the implementation of such computer processing of experimental relaxation dependencies is presented, which makes it possible to implement the choiceof

- the most informative time interval from the entire measurement time range for the processed kinetic dependencies in order to most accurately estimate all model parameters;

- optimal analytical expressions for representing trends in the influence of one or another specified external factor on the parameters of the specified function.

The proposed algorithm and its software implementation are used to process research data on gas-sensitive sensor materials. The results of the testing of the developed application showed satisfactory correspondence of the obtained results to the existing physical concepts and allowed us to conclude that this approach is promising for automating the processing and analysis of experimental relaxation dependencies.

Keywords: data processing, information-measuring system, relaxation dependence, stretched exponential function, gas-sensitive sensor, response relaxation.

Received 25.09.2024; Received in revised form 15.10.2024; Accepted 12.11.2024

1. Introduction

The study of relaxation processes is currently the most effective approach for obtaining the most complete information about the properties of a particular object. Analysis of kinetic dependencies is one of the most common methods for studying a wide range of relaxation processes in the field of natural sciences, such as global earthquake systems, galactic light radiation, economic phenomena, etc. [1-3].

Ensuring sufficient accuracy and information content of the results obtained with this approach involves the need for many measurements and repeated analysis of the results obtained. As a rule, this takes a lot of time and requires many cycles of processing large amounts of experimental data. To eliminate routine operations associated with the processing of experimental relaxation dependencies; to increase the speed, accuracy, and information content of the research process itself, it seems relevant to automate it using computer technology.

One of the options for constructing hardware and software for a specialized automated information-measuring system (IMS) that would provide digitization, input of measurement data into memory and data analysis within the framework of the well-known model of a stretched exponential function is considered in [4]. The stretched exponential function (StrEF), also known as the Kohlrausch-Williams-Watts function, is one of the most common phenomenological models for describing relaxation processes: $f(t) = \exp[-(t/\tau)^{\beta}]$ where t is time, β and τ – parameters [5-6].

The main feature of this model is the possibility of its multi-purpose application to solve various problems:

- presentation of experimental data in an analytical form that is more convenient for analysis;

- obtaining on its basis information about the spectral characteristics of the relaxation processes under study, which is based on its mathematically established connection with the phenomenological model that describes the frequency properties of the relaxing system, for example, the Cole-Cole formulas [7] for dielectrics;

- interpretation of StrEF parameters and their dependencies on various factors in physical models of relaxation processes when studying the properties of various materials and technical systems.

In materials science, StrEF is widely used to explain and describe the behavior of glassy states, luminescence decay and other electronic systems from the point of view of the distribution of relaxation times [1, 8-13]. Recently, such a model has been used to describe the kinetics of relaxation dependences of the response of gas sensors at the stage of restoration of the original electrical conductivity [13]. As shown in [14-15], the use of this approach makes it possible to obtain significantly more information about the physicochemical phenomena that determine the phenomenon of gas sensitivity of sensor materials. The data presented in [4] indicates the reality and effectiveness of computer processing of a significant amount of data in the process of carrying out the measurements under consideration.

The main tasks, the implementation of which was considered, were the following:

- digitization and input of measurement data into the computer memory in the form of text files;
- processing and analysis of data within the framework of the previously mentioned stretched exponential function model, in particular, determination of parameters and construction of their dependencies on various factors (temperature, partial pressure of the detected gas, design features and chemical composition of the sensor used, etc.).

The first of the data processing algorithms used in this work does not allow the implementation of a fully automated IMS for studying the characteristics of gas-sensitive sensors. The main reason for this is the need to numerically calculate derivatives and select the optimal interval for their calculation from the time dependence of the sensitivity (response) of the sensor to the active gas. Performing such intelligent actions requires direct participation in the operator's data processing process.

The second algorithm used, described in [16], eliminates the participation of the operator in processing the measurement kinetics data. However, in this method, the value of the amplitude parameter S_0 of such a stretched exponential function must be determined independently, which is not always possible.

It should be note that one of the rapidly developing areas of ensuring automation of computer processing of measurements is the use of artificial intelligence tools and methods [17]. These methods are used in systems for monitoring the physical properties of various materials [18-19], measuring mechanical quantities (for example, the position of aircraft) [20-21], testing and analyzing faults of electronic devices [22], etc. Application that seems promising approaches and means for implementing artificial intelligence methodology to improve software, more fully automate processing and primary analysis of the relaxation dependencies is considered here.

This paper presents the results of developing and testing software using a stretched exponential function based on the addition of the first of the mentioned algorithms with intelligent components that eliminate operator participation in the process of processing and primary analysis of measurement data using the example of response relaxation of resistive gas sensors.

2. Methodology for analyzing relaxation dependencies using the stretched exponential function model

The advantage of the algorithm for processing and primary data analysis used in [13, 14-15] is that there is no need for any assumptions or additional data. Only the results of measurements of the kinetic dependence of the response of a gas-sensitive sensor $S(t)$ are used. The corresponding approximating dependence is used as a model for the primary phenomenological analysis of the experimental kinetic dependences S(t). After removing the active gas from the surrounding atmosphere, it was assumed to be represented in the form

$$S(t) = S_0 \cdot \exp\left[-(t/\tau)^{\beta}\right] . \quad (1)$$

In accordance with [13, 23], the mathematical procedure for finding the value of the unknown coefficient β includes the following operations:

- numerical determination of the derivatives $d\lg S(t) / dt$ based on the experimental kinetic dependence of the form (Fig. 1) and representation of this dependence in coordinates $t\times[d\lg S(t) / dt]$ and $\log S(t)$;

- visual selection (by the operator) of the area above the specified dependence, where the model is applicable (i.e., the dependence becomes straight, and the coefficient β itself is the tangent of its angle) and calculation of the value

$$\beta = \Delta\{t\times[d\lg S(t) / dt]\}/\Delta \lg S(t). \quad (2)$$

Representation of the initial relaxation dependence in the selected area in coordinates $\ln S(t)$ and t^{β} (β is already known), i.e. approximation of this dependence by a straight line allows you to determine the remaining parameters StrEF (1) using the following formulas

$$\tau = [-\Delta \ln S(t) / \Delta (t^{\beta})]^{-1/\beta}; S_0 = \exp\left[\ln S(t) + (t/\tau)^{\beta}\right]. \quad (3)$$

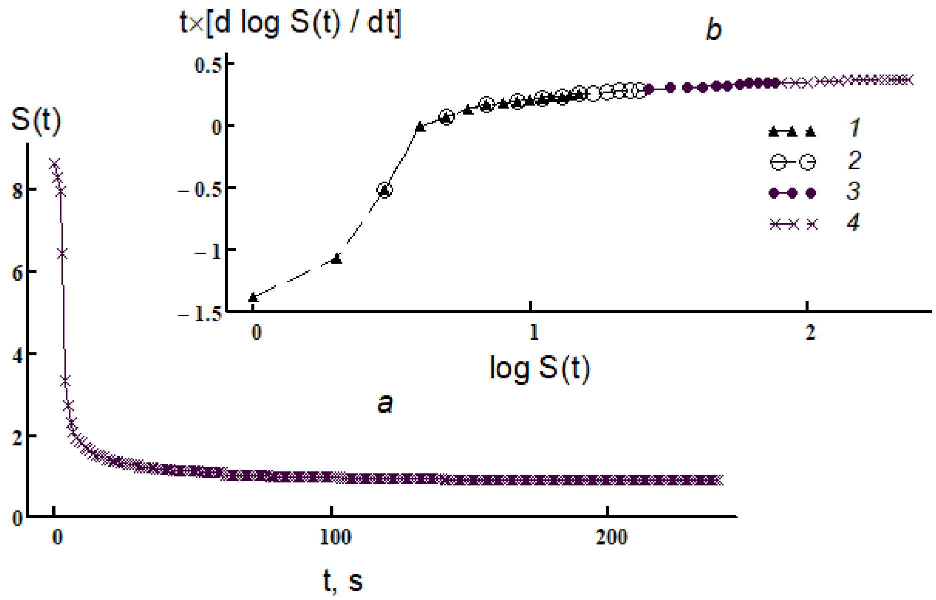

Fig. 1. Relaxation curve of the response of a gas sensor sample upon restoration of its initial state in different coordinates (a). Selected time slots are indicated by numbers 1, 2, 3 and 4 (b).

3. Algorithm for processing relaxation dependencies

In accordance with the above description of the method under consideration, one of its steps is an intellectual operation – the selection of the most suitable section of the relaxation dependence presented in Fig. 1b for calculating the parameter β. As can be seen, in a real situation the model can describe relaxation processes with different levels of approximation. Thus, in the general case, the task of determining the optimal value of the parameter falls on the actions of the operator accompanying the data processing. One of the ways to implement these intellectual actions may be to divide the entire time range of relaxation into several parts – intervals (permissibly overlapping); defining sets of StrEF parameters for each such interval; choosing the one that provides the smallest error.

A description of the main operations of the corresponding algorithm can be present as follows.

1. Divide the entire time range of each (k-th) experimental dependence $S(t)$ into several intervals with a sufficient number of points for the numerical determination of the time derivative. These intervals are considered independent and are used to calculate several (m) sets of values for each of the parameters of the approximating StrEF (β, S_0, τ), which can be presented in the form of tables $\{\beta_{k,j}\}$, $\{S_{0k,j}\}$ and $\{\tau_{k,j}\}$, respectively, here k is the index of the relaxation curve, which corresponds to the k-th value of the factor, which influence is studied in this experiment (for example, temperature T). Index k varies from 0 to (m –1), where m is the number of processed dependencies corresponding to different factor values. Index j is the number of the interval of a separate processed dependence, varies from 1 to n (in the example given n = 4).

It should be noted that the parameter values (β, S_0, τ), found in different time intervals have some scatter, the presence of which can be associated with the influence of both a random factor and the general degree of approximation of the description of relaxation processes of this type using StrEF (Fig. 1b).

2. For each interval of the experimental relaxation dependence, determine the coefficients of the approximating expression of the form (1) using the well-known least squares method [24], the minimized function for which can be write in the following form:

$$\Phi_{kj} = \sum_{i=0}^{N_k-1}\left[\lg\left(S_k^i\right) - \lg\left\{S_{0kj}\cdot\exp\left[\left(t_k^i/\tau_{kj}\right)^{\beta_{ki}}\right] + \delta\right\}\right]^2 , \qquad (4)$$

where t_k^i and S_k^i are i-th value of time and response of the processed experimental relaxation dependence ($N_k$is the total number of experimental points of the k-th processed relaxation dependence).

The use of logarithmic expressions in (4) is due to the wide range of changes in the values of the relaxation functions (from infinitely large to infinitely small values). Parameter δis a small positive value ($\delta \sim 10^{-15}$), which allows eliminating the mathematical uncertainty of the logarithm of the approximating function at small values.

The parameters $\tau_{k,j}$ and $S_{0k,j}$ are determined by presenting experimental relaxation dependencies in coordinates ln S_k^i and $\left(t_k^i\right)^{\beta_{k,j}}$in each j-th section of them with a straight line and then using calculation formulas (3) [13, 23].

To select the optimal values of the parameters β, S_0 and τ of the k-th dependence of the form (1), the condition for the minimum value of the corresponding root-mean-square error of approximation was used, that is, the condition

$$\text{IF } \Phi_{k,j_{min}} = \min\left(\Phi_{k,j}\right),\ \text{THEN } j_{min}\text{-th set of parameter valuesis selected.} \qquad (5)$$

To improve the quality of approximation, the size and location of the selected intervals can be changed using other logical algorithms. Intervals can be extended or shifted.

It should also be noted that the overall optimality of the approximation could be assessed using the coefficient of variation (the standard deviation of the approximation divided by the empirical mean):

$$\mathrm{var}_k = \sqrt{\frac{1}{N_k - 1} \cdot \Phi_{k,j}} \Big/ \sum_{i=0}^{N_k-1} \frac{\lg(S_k^i)}{N_k}$$

3. The values of parameters β_k, S_{0k} and τ_k selected in this way, optimal for each relaxation dependence, used to analyze the patterns of influence on it of one or another factor, the changes of which specified in a particular study.

4. Analytical representation of the dependences of StrEF parameters on various factors

The choice of the type of formulas that approximate the dependences on the parameters $\beta(x)$, $S_0(x)$,and $\tau(x)$ of StrEF (1) on the given factor x can be based on dividing them into two most general classes: monotonic (decreasing or increasing) and having extremum. It assumed that in the studied, usually limited, range of factor values, monotonic ones can be approximately described by the simplest regression equations of the form

$$\beta_1(x, a) = a_0 \cdot \exp(-a_1 \cdot x), \tag{6}$$

$$\beta_2(x, a) = a_0 + a_1 \cdot x + a_2 \cdot x^2, \tag{7}$$

and having an extremum by the formula

$$\beta_3(x, a) = a_0 \cdot \exp\left[-\frac{(x - a_1)^2}{2 \cdot a_2{}^2}\right]. \tag{8}$$

If necessary, a number of approximating functions can be expanded and changed. The selected approximation formulas for different StrEF parameters taken from their common pool (i.e., formulas (6)–(8) in this case).

The values of the coefficients a_0, a_1 and a_2 for the formulas found using the least squares method, i.e. by minimizing the function

$$F_l(a) = \sum_{k=0}^{n-1}[\beta_k - \beta_l(x_k, a)]^2 \tag{9}$$

where l= 1, 2, 3.

An analysis of the level of their suitability for approximating the dependencies of tabular data obtained using the method described in the previous section was carried out. To do this, the coefficients of regression dependencies (6) – (8) and the corresponding coefficients of variation $\mathrm{var}_l^{(z)}$ calculated using the least squares method:

$$\mathrm{var}_l^{(z)} = \sqrt{\frac{F_l(a)}{m-1}} \Big/ \sum_{k=0}^{m-1} \frac{\beta_k}{m}.$$

The notation z takes the values β, S_0 and τ, respectively. The final choice of the approximating formula was justified in accordance with the simplest production rule of the form (5):

IF $\mathrm{var}_{lm}^{(z)} = \min(\mathrm{var}_l^{(z)})$, THEN the lm-th approximating formula is selected. (10)

5. Software implementation

The environment that ensures the operation of the described software product is the Windows operating system. Navigation between its sections in the simplest case considered here is implemented using buttons [25].

To control the processing of source data, the Batch Monitor is used, which written using the C# language and the Windows Forms Application project type of the Visual Studio environment. Application software modules of the library of calculation algorithms are documents of the well-known mathematical package Mathcad (version 15) [26]. In addition, the application uses universal tools for working with files with extensions such as .xlsx. (or/and .dat).

The user interface takes into account that this application is based on the integrated use of heterogeneous software products and provides the following service functions for processing and analyzing the specified data (Fig. 2):

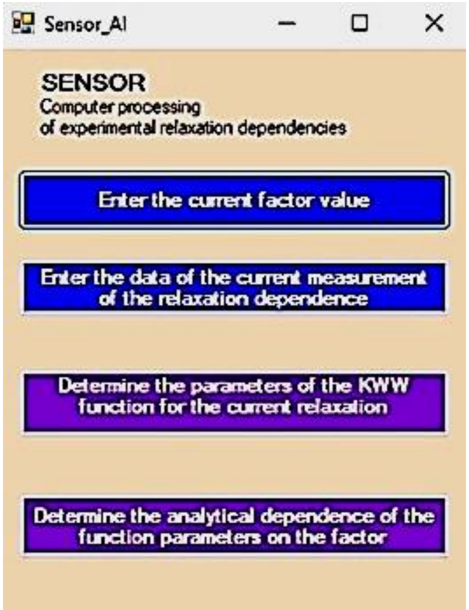

Fig. 2. Main application user interface window

1. Data entry.

The following are loaded into Excel spreadsheet files:

- the value of the factor whose influence is being studied in accordance with the program of a specific study (go to the window of the first file (parameter file) – click on the button “**Enter the current factor value**”),

- a table of experimental data of the relaxation dependence corresponding to this factor value (go to the window of the second file (data file) – click on the button “**Enter the data for the current measurement of the relaxation dependence**”);

2. Calculation in the mathematical package "Mathcad" of the StrEF parameters for a given factor value and transferring them to the mentioned parameter file (go to the window of the first software module of the "Mathcad" package – click on the button "**Determination the parameters of the KWW function for the current relaxation**");

3. After accumulating the data corresponding to the used values of the specified factor in the parameter file, clicking on the button “**Determination of the analytical dependence of the function parameters on the factor**” ensures the transfer of the specified data to the

second software module of the mathematical package “Mathcad” for determining the dependence of the StrEF parameters on the factor in question in an analytical form, including their visualization.

6. Test results

The main characteristics of gas-sensitive sensors are [27]:

- response kinetics when a certain amount of adsorbent enters/removes into the atmosphere surrounding the sensor (at a constant temperature of the sensor);
- temperature dependence of the response at a constant adsorbent concentration.

The response was determined as R_0/R_S, where R_S is the resistance of the sensor in a medium containing adsorbite, R_0 is in air. The measuring setup described in [4] was used. It consisted of a sealed chamber with a volume of 20 dm^3 to which a small expansion tank made of elastic rubber was attached to equalize the pressure between the volume of the chamber and the surrounding atmosphere, a power supply, an electrical circuit that ensured the reception of a signal from the sensor and its transmission to a personal computer (PC), and the computer itself. The gas sensor, together with the system for ensuring its heating, was in the measuring chamber and was electrically connected to the rest of the installation. The chamber also contained a thermometer to control the temperature of the gases and a miniature fan to ensure the isotropy of the gas environment.

A dosed amount of active gas was introduced at the right moment into the measuring chamber. The effect of temperature on the response kinetics of ZnO-Ag ceramics in ethyl alcohol (C_2H_5OH) vapor studied.

The results of testing the IMS considered for computer processing of measurement data of gas sensor parameters in automatic mode are shown in Fig. 3.

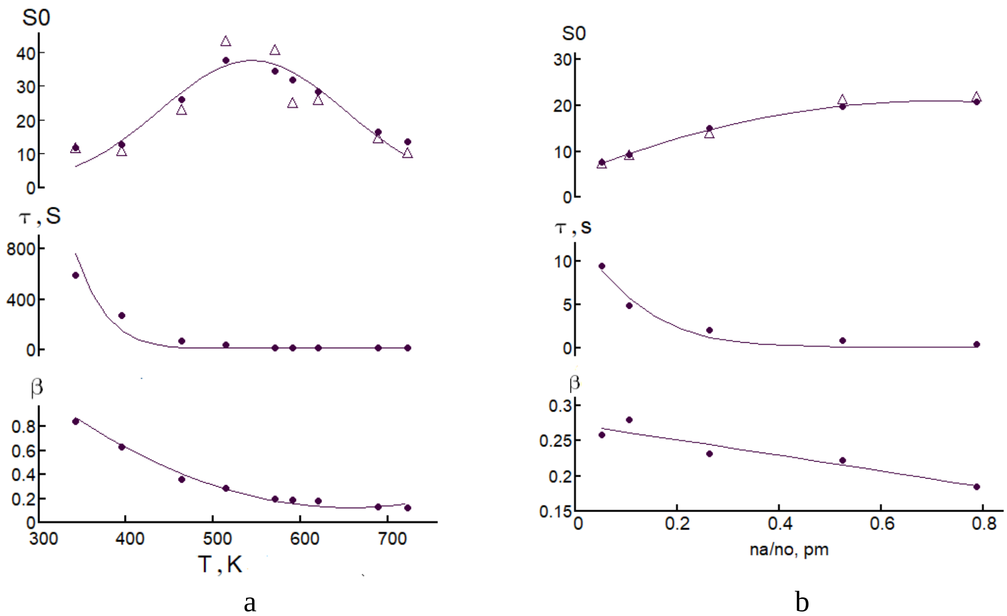

Fig. 3. Dependences of the stationary (maximum) value of the response S_0, the characteristic time τ and the elongation index of the duration of this process β of a sample of zinc oxide ceramics with the addition of silver (2 wt. %) on temperature at a relative concentration of ethyl alcohol vapor in the air of 0.26 pm) (a) and on the relative concentration of ethyl alcohol vapor (n_a/n_o) at a temperature of 685 K (b). The dots are the result of processing, the solid line is the result of approximation by an analytical expression, the triangles are the results of direct measurement of the static value of the response.

The presented results indicate the presence of a maximum sensitivity temperature of the sensor (close to 550 K), a decrease in the characteristic time and duration of the relaxation process.The presented results indicate the presence of a temperature of maximum sensitivity of the sensor (close to 550 K), a decrease in the characteristic time and duration of the relaxation process with increasing temperature T and n_a/n_0 concentration, which corresponds to existing ideas and specific literature data [28]. The measurement data of the static (maximum) response value shown in this figure, as can be seen, is also in satisfactory agreement with the results of the used algorithm for processing experimental relaxation dependencies.

Conclusions

An option is present for implementing computer processing of experimental relaxation dependencies based on a model of a stretched exponential function, which uses intelligent components that make it possible to implement the choice

- the most informative time interval from the entire measurement time range for the processed kinetic dependencies in order to most accurately estimate all model parameters;

- optimal analytical expressions for representing trends in the influence of one or another specified external factor on the parameters of the specified function.

The considered algorithm and its software implementation used for computer data processing using the example of the study of gas-sensitive sensor materials. The results of the testing of the developed application showed a satisfactory correspondence of the obtained results to the existing physical concepts and allowed us to conclude that this approach is promising for automating the processing and primary analysis of experimental relaxation dependencies.

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