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INTERVAL MODEL FOR ASSESSING THE PURITY OF NUCLEAR STRUCTURAL METALS BY MICROHARDNESS

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The article considers the possibilities of evaluating the purity of Zr-1%Nb zirconium alloy samples – the main structural material of the thermal neutron reactors active zones with a water coolant – according to the microhardness of the metal. The correctness of the application of interval regression analysis is shown, if there is uncertainty, incompleteness of information, measurement errors in the experimental data for determining the oxygen content in metal samples and its microhardness. The method of estimating parameters of interval linear regression is presented.

INTRODUCTION

The central part of the nuclear reactor is the active zone. In it, the processes of fuel division, energy release, and removal of thermal energy by the coolant take place. The active zone is the most stressed, responsible, and vulnerable part of the reactor facility. In the active zone, structural materials work in extreme conditions. These conditions are intensive neutron and other irradiation, the influence of the heat carrier of high parameters and fission products, and thermomechanical loads.

The role of structural materials consists in ensuring the stability of the geometry of the active zone for the entire period of operation, primarily of heat-emitting elements and heat-emitting assemblies, in keeping fuel fission products inside the heating element, maintaining the efficiency of control and protection bodies, ensuring the minimum possible consequences of emergencies, i. e. in solving of the important safety issues of the reactor facility [1].

The primary structural material of the active zones of thermal neutron reactors with a water coolant are zirconium alloys – E110, E635, E125, M5, Zry-4, and ZIRLO [2, 3]. Today, the underlying material of the fuel shells of active Ukrainian reactors remains the E110 alloy (Zr-1%Nb), the principal alloying element of which is niobium. Among the promising foreign zirconium alloys also doped with niobium (with different percentages of this metal) are ZIRLO and M5 (1.0 wt.%), NDA (0.1 wt.%), MDA (0.5 wt.%). The optimal combination of nuclear, corrosion, mechanical, thermal, and other physicochemical characteristics make zirconium alloys indispensable structural materials for nuclear energy.

About 20 impurities in zirconium alloys for nuclear reactors are regulated. The effects of impurities on the properties of alloys, as well as their structure, are diverse. Gas-forming elements make a decisive contribution to the value of the total content of impurities: their average content in metal samples is at the level of 10^{-4} at.%. A further increase in the purity of metals is achieved by removing the main gas-forming impurities. The development of methods for removing oxygen from zirconium is urgent, because the increased gas content in a zirconium alloy with niobium worsens its technological properties.

The purity of the metal has a significant effect on the microstructure and mechanical properties of zirconium. In particular, microhardness significantly depends on the concentration of impurities in the metal, and according to the data of observations and measurements, it forms a linear dependence.

Thus, it is possible to draw conclusions about the purity of the material based on the change in the microhardness value and, accordingly, estimate the amount of impurities.

A large number of scientific and research works are devoted to the determination of oxygen content in zirconium alloys. Almost all works emphasize that the obtained results are often underestimated and unreproducible (the reason is the unequal conditions of oxygen release in a series of experiments). Gases absorbed by zirconium affect its brittleness and inability to be machined under pressure. Namely - metal forging, and rolling, the strength limit increases, and the plastic properties decrease significantly.

Since the middle of the last century, scientists have considered various methods for determining the oxygen content: the vacuum melting method (there is a melting of a metal sample in a previously degassed vacuum system in the presence of an excess of carbon; then the gases that are released are pumped out using a system of pumps and subjected to analysis), isotope method, chlorination method, vacuum extraction method, platinum flux method, emission spectral analysis method, application of high-temperature induction heating of the sample or deoxidizing platinum baths, electron energy loss spectrometer, X-ray energy dispersive spectrometer, etc. [4–13].

The analysis of literature data on the error and sensitivity of the methods shows that the error of the determined amount of oxygen is 5…6%, when the gas content in the alloy samples is 10^{-2}…1 wt.%, and accordingly 10…12% – for $(2\cdot10^{-3})$…$(5\cdot10^{-2})$ wt.% oxygen content.

The work [14] gives a formula: the calculation method determines the required oxygen content in zirconium alloy samples with dosed surface oxidation followed by annealing in a high vacuum to dissolve oxygen inside the sample. The weight of the sample and its increase for calculation is determined. The error of each measurement is from 0.4 to 2%. The total error when calculating the oxygen content according to the formula is up to 5%.

It was clear that the specified methods and approaches for determining the oxygen content are expensive, not elementary, and have their sensitivity limits. Determining the amount of gas impurities, especially oxygen, in zirconium alloys is significant and an actual task. Oxygen, on the one hand, increases the strength of zirconium, and, on the other hand, when combined with other impurities, it negatively affects the structure and properties of the alloy. Therefore, obtaining a value as close as possible to the true one is an important issue.

The existing generalized data application on the hardness measurement and the dynamics of changes in the oxygen content (and other impurities) in zirconium alloy samples based on the results of electron beam melting of the metal in a vacuum is a helpful approach for solving this problem. We determine the parameters of impurities, that is, the purity of the alloy, based on the hardness data.

Microhardness and its measurement accuracy are important for studying the mechanical properties of zirconium materials. Traditionally, PMT-3 microhardness tester to determine microhardness is applied. The standard device error of the according is 2%. The nanoindentation technique to study the mechanical properties of thin (thickness of several microns) layers of the surface of materials is applied. This technology does not use the size of the impression area to determine the hardness of the alloy sample.

We come to the conclusion that the experimental data resource, which will allow us to evaluate the quality of the alloy sample, its purity, mechanical properties and, finally, to build models of dependencies between various indicators and the zirconium alloy properties contains uncertainty, which is a consequence of many factors: unequal conditions of conducting research, instrumental measurement errors, methodological errors due to limitations of methods and inaccuracy of approximate calculations, ambiguity of results, chaotic distortions of an unknown quantity, etc. There is no guarantee that the observations have the same accuracy. In addition, the probabilistic characteristics of all error components are unknown or not Gaussian distribution; the limit on the maximum value of the cumulative errors is also uncertain. In such a situation, the treatment of classical regression analysis approaches to determine the correlation between measured values, regression parameters, and the type of regression dependence itself is not entirely correct.

Therefore, the methods of interval analysis and interval statistics [15, 16] are becoming increasingly popular, in which the basic concept is the set of measurement uncertainty, which is guaranteed to contain the true value of the observation.

In previous works, the authors of the article considered the application of this mathematical apparatus for assessing the safety and reliability of the systems and equipment of the NPP power unit [17], building operational and temperature characteristics of the separation system [18], modeling the properties of nuclear structural materials [19–22].

In [19] authors proposed the Interval model. It is constructed on the basis of experimental data, based on the following assumptions: the value of the oxygen content is reliably known, and the measured values of the microhardness of the alloy samples contain errors and are specified using intervals. In other words, input data are precise (point), and output data are interval values. The model was sought in the form of a linear function

$$\mathbf{Y}(x) = \mathbf{b_1} + \mathbf{b_2} \cdot x\,, \tag{1}$$

where $\mathbf{Y}(x)$ – microhardness of the alloy sample; x – oxygen content in the sample; $\mathbf{b_1}, \mathbf{b_2}$ – required dependence parameters (interval values).

The results of the simulation and the model parameters values are presented in Figs. 1, 2.

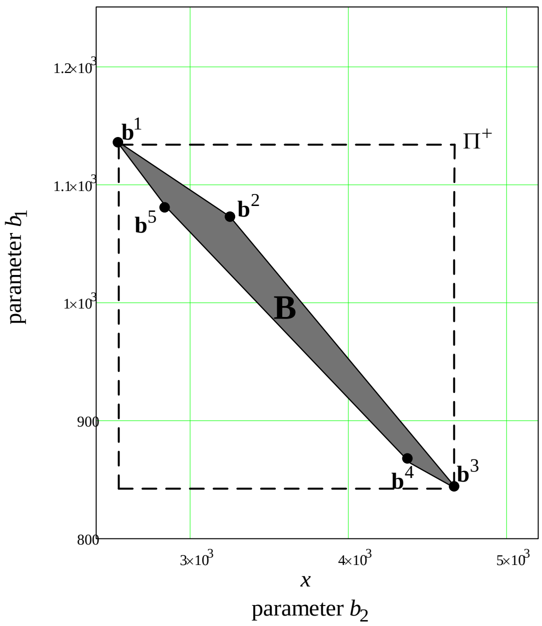

Fig. 1. The membership set $\mathbf{B}$

Set of model parameters $\mathbf{B}$ (see serial pentacle, Fig. 1) – the solution of the linear programming task – task (1) from interval experimental data on the output unknown $\mathbf{Y}(x)$. The extreme cut points $\mathbf{b^1}$ and $\mathbf{b^3}$ indicate the size of the multiplier B and, apparently, the rectcutaneous point, which is an approximation of the model parameters set (1):

$$\mathbf{\Pi^+} = \{(b_2, b_1) : 2538 \le b_2 \le 4750, 836 \le b_1 \le 1137\}.$$

And, if instead of the set $\mathbf{B}$ itself, its approximation estimate is applied, then the corresponding interval model will have the form:

$$\mathbf{Y}(x) = [836;1137] + [2538;4750] \cdot x\,.$$

Fig. 2 shows a tube (lines 1) – a set of permissible values of dependence (1). That is a forecast model. Model contains all possible values of the output variable with different choices of dependence parameters it constructed on the basis of the set of uncertainties of parameters $\mathbf{B}$.

Point estimates of the model presented in Fig. 2, demonstrate the dependence constructed by the least squares method (line 2), which passes outside the uncertainty intervals of the observations, the dependences whose graphs practically coincide (line 3 and line 4) – estimations of the parameters of the forecast model are calculated by the method of the uncertainty center and the gravity center of the set $\mathbf{B}$.

The proposed interval model and work with interval data make it possible to avoid the loss of information, which is a consequence of the processing of purely single-valued measurement values. But you should pay attention to the following. Analysis of the estimation of the set of dependence parameters (1) shows that the approximation rectangle $\mathbf{\Pi}^+$ (see Fig. 1) also contains extraneous values. Moreover, the set $\mathbf{B}$ is significantly different from a rectangle $\mathbf{\Pi}^+$; the polygon $\mathbf{B}$ has a very shape elongation.

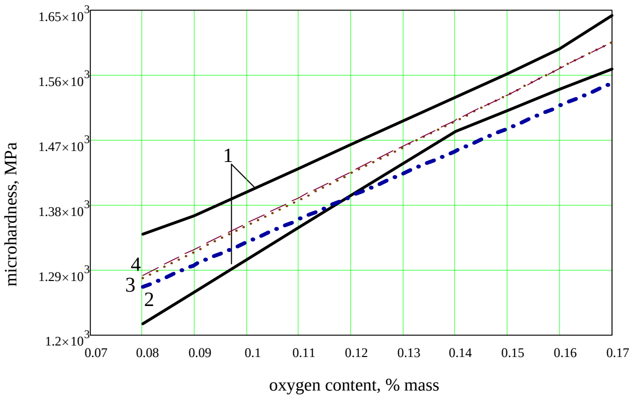

Fig. 2. Interval and point estimates of the forecast model

Recently, various scientists have often asked questions – of how the processing and interpretation of experimental data is affected not only by the uncertainty of the data itself but also by the inaccuracy of the method used to estimate the parameters. These reasons led to the development of new statistical analysis methodologies for processing variables with interval values [23–26] and, the construction of interval regression dependencies [27–29].

In [25], the authors F. Gioia and Carlo N. Lauro introduced basic statistical indicators for interval variables, proved the properties that correspond to single-valued values of the variables, proposed a method for selecting a simple interval linear regression equation.

This approach allows you to build a forecast model - a linear dependence of variable Y with interval values on one independent variable X with interval data.

The analysis of the methods for determining the oxygen content in zirconium alloys confirms that the point values of the oxygen content in the metal samples are not actual and taking into account measurement errors, method errors, etc., it is correct to consider interval data.

Thus, the dependence model constructing the purity of Zr-1%Nb zirconium alloy samples on hardness is a task with interval input and output data.

The method idea from work [25] is to consider all possible values of components x_i, y_i so that each is in its range (intervals) of values $X_i = \left[\underline{x_i}, \overline{x_i}\right]$ and $Y_i = \left[\underline{y_i}, \overline{y_i}\right]$, $i = \overline{1,n}$ to construct an interval linear regression. For different sets of values selected from each interval, the set of "interval points" and the calculation of the corresponding regression (unambiguous two parameters of the linear function) are uniquely determined (Fig. 3). Task: finding the set of parameters of all possible regression lines for different sets of x_i, y_i values, $i = \overline{1,n}$.

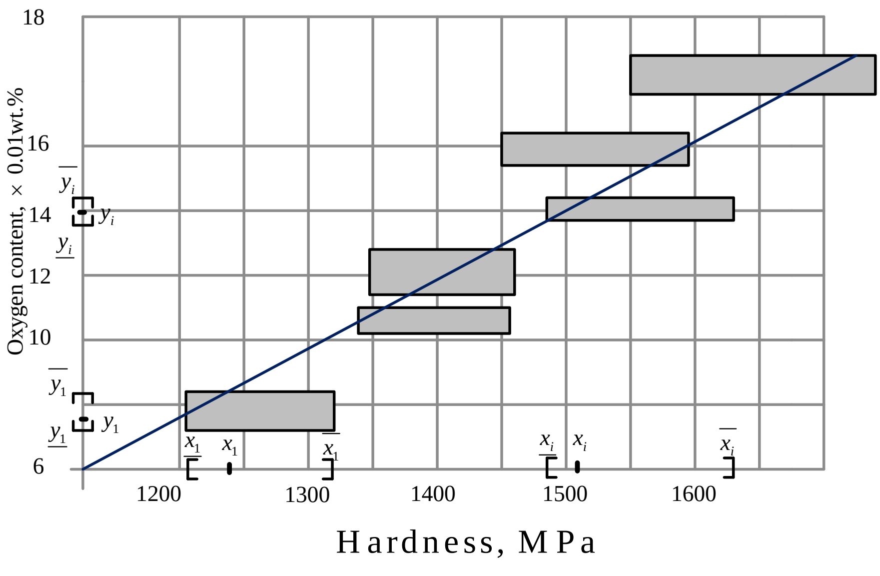

Fig. 3. A set of "interval points" is constructed for one arbitrary values x_i, y_i, $i = \overline{1,n}$

Thus, constructing a regression between two interval variables means determining a set of regression lines, each of which realizes the best correspondence, in the sense of the Least Squares Method, to a set of points on the plane. This set of points is uniquely determined whenever the components $x_1,...,x_n,y_1,...,y_n$ are assigned a value from their variation intervals.

Interval regression has the form:

$$y = \hat{\boldsymbol{\beta}}_0^I + \hat{\boldsymbol{\beta}}_1^I \cdot x \,, \tag{2}$$

where

$$\hat{\boldsymbol{\beta}}_0^I = \left[\min_{\substack{x_i \in [\underline{x_i}, \overline{x_i}] \\ y_i \in [\underline{y_i}, \overline{y_i}]}} \hat{\beta}_0, \max_{\substack{x_i \in [\underline{x_i}, \overline{x_i}] \\ y_i \in [\underline{y_i}, \overline{y_i}]}} \hat{\beta}_0 \right], \tag{3}$$

$$\hat{\boldsymbol{\beta}}_1^I = \left[\min_{\substack{x_i \in [\underline{x_i}, \overline{x_i}] \\ y_i \in [\underline{y_i}, \overline{y_i}]}} \hat{\beta}_1, \max_{\substack{x_i \in [\underline{x_i}, \overline{x_i}] \\ y_i \in [\underline{y_i}, \overline{y_i}]}} \hat{\beta}_1 \right], \tag{4}$$

functions

$$\hat{\beta}_1\left(x_1,...,x_n,y_1,...,y_n\right) = \frac{\sum_{i=1}^{n}\left(x_i - \overline{x}\right)\left(y_i - \overline{y}\right)}{\sum_{i=1}^{n}\left(x_i - \overline{x}\right)^2}, \tag{5}$$

$$\hat{\beta}_0\left(x_1,...,x_n,y_1,...,y_n\right) = \overline{y} - \hat{\beta}_1 \overline{x} \,, \tag{6}$$

in addition

$$\overline{x} = \frac{1}{n}\sum_{i=1}^{n} x_i,\ \overline{y} = \frac{1}{n}\sum_{i=1}^{n} y_i \,,$$

$$x_i \in \left[\underline{x_i}, \overline{x_i}\right], y_i \in \left[\underline{y_i}, \overline{y_i}\right],\ i = \overline{1,n} \,.$$

The authors introduced concepts of interval means, deviation from the mean, interval variance, covariance, and correlation interval matrices [25]. They allow us to find out whether the experimental data are well correlated and what the expected forecast model might be.

CONCLUSIONS

A review of existing methods for determining the content of gas impurities, which significantly affect the structure and properties (in particular, microhardness) of the Zr-1%Nb zirconium alloy, showed that it is advisable to use models for assessing the purity of alloys based on microhardness indicators. Regression between two interval variables is a model that considers uncertainties in data, incomplete information, measurement errors, and errors in parameter estimation methods. The methodology of interval statistics also allows you to determine how close the relationship is between the values of measured experimental data.

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ІНТЕРВАЛЬНА МОДЕЛЬ ОЦІНКИ ЧИСТОТИ ЯДЕРНИХ КОНСТРУКЦІЙНИХ МЕТАЛІВ ПРИ МІКРОТВЕРДОСТІ

Т.В. Потаніна, О.В. Єфімов, М.М. Пилипенко

Розглянуто можливості оцінки чистоти зразків цирконієвого сплаву Zr-1%Nb, який є основним конструкційним матеріалом активних зон реакторів на теплових нейтронах з водним теплоносієм, за показниками мікротвердості металу. Показано коректність застосування інтервального регресійного аналізу в ситуації невизначеності, неповноти інформації, похибок вимірювань в експериментальних даних визначення вмісту кисню в зразках металу і його мікротвердості. Представлено метод оцінки параметрів інтервальної лінійної регресії.