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THE METHOD FOR OPTIMIZING THE IRON CONTENT IN THE STRUCTURAL MATERIAL Zr1%Nb FOR FUEL ELEMENT CLADDING OF NPP NUCLEAR REACTORS

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The mathematical method of optimizing the amount of the alloying element Fe in structural zirconium alloys Zr1%Nb of fuel elements cladding of nuclear reactor-cores of nuclear power plants on the basis of physical experiments to increase their corrosion resistance is considered. Alloying the Zr1%Nb alloy with Fe is promising in the development of the technology for the production of domestic materials for fuel elements claddings for reactors with high reliability and safety. To process the results of experimental studies of corrosion of zirconium alloy with different Fe content, a mathematical method of two-dimensional polynomial comb regression was proposed with its implementation in the Python programming language based on the theory of "machine learning". The application of this method made it possible to determine the optimal amount of the alloying element Fe for zirconium alloy Zr1%Nb of fuel elements cladding of nuclear power plant reactors with pressurized water.

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INTRODUCTION

The main structural materials for fuel element cladding of water-cooled thermal neutron reactors both in our country and abroad are zirconium alloys, which, unlike other structural materials, have a very small thermal neutron absorption cross-section (0.18 barn), which is necessary for the use of low-enriched nuclear fuel. Such zirconium alloys used in the nuclear power industry of various countries (Ukraine, Canada, USA, France, Japan, etc.) include alloys Zircaloy-4 (Zry-4), E110, E635, E125, M4, M5, MDA, and others. In the USA, Canada, and Western Europe, two main zirconium alloys are used for fuel element cladding, shrouds and channels of light-water and heavy-water reactors: Zircaloy-4 and Zircaloy-2, and the first is mainly used for fuel element reactors PWR, the second – for reactors BWR [1, 2].

In Table 1 shows the chemical composition and mechanical properties of some zirconium alloys. Comparison of the mechanical properties of Zry's and Zr-Nb alloys shows the difference of the zirconium alloys in both strength and elongation.

In the programs of work carried out in the world to improve nuclear fuel for new-generation WWER (PWR) reactors and which are aimed at further improving the operational reliability of fuel cells with fuel burn-up to 70…75 (GW·days)/t U) and a campaign length of up to 6…7 years, much attention is paid to increasing the resource characteristics of zirconium fuel element cladding and components of fuel assemblies.

One of the most important requirements for the structural materials of nuclear reactor cores is their high corrosion resistance, which should ensure the reliability and safety of operation of both the reactor plant and the entire power unit as a whole. Meeting the corrosion resistance requirement is particularly important for structural materials of reactor fuel element shells, which are made of Zr1%Nb zirconium alloy. With a minimum wall thickness (0.65 mm) for neutron absorption, they are in the most difficult operating conditions in the core. That is, during operation, the material of fuel element shells can deform as a result of radiation damage to the nuclear fuel in the fuel element, large thermal stresses occur in the cladding due to significant temperature changes, and the material of the cladding can change its physical and mechanical properties under the influence of neutron irradiation. Therefore, ensuring high corrosion resistance of fuel element cladding of nuclear reactors of nuclear power plants is an urgent scientific and technical problem of the world nuclear power industry [1, 3].

The results of long-term studies of the purification processes of zirconium and alloy Zr1%Nb, which was obtained using Ukrainian technology in the form of ingots in the NSC KIPT of the National Academy of Sciences of Ukraine, are presented in [3–6]. Various methods in laboratory conditions were used for those researches, and properties of the alloys were determined using mathematical methods for processing experimental data, taking into account the uncertainty of the initial data. Research on the structure, chemical composition, thermal desorption, hardness and microhardness of Zr1%Nb alloy samples, as well as its corrosion resistance were carried out.

Zirconium alloy Zr1%Nb, in which Nb is an alloying element, contents of about 20 impurities (Hf, Al, Ti, Fe, N, F, C, Si, Cl, etc.). At the same time, it is known that Al, Ti, N, C reduce the corrosion resistance of zirconium in water at high temperatures, and Fe increases the corrosion resistance of zirconium in water and steam environments [7].

Table 1

Chemical composition of zirconium alloys [1, 3]

Parameter	Zircaloy-2	Zircaloy-4	E110	E125
Chemical composition, wt.%				
Zr	98.6…97.8	98.4…97.8	~99.0	~97.5
Nb	–	–	1.0	2.5
Sn	1.2…1.7	1.2…1.7	–	–
Fe	0.05…0.15	0.18…0.24	–	–
Cr	0.07…0.20	0.07…0.13	–	–
Ni	0.03…0.08	–	–	–
Σ (Fe+Cr+Ni)	0.18…0.38	–	–	–
Σ (Fe+Cr)	–	0.28…0.37	–	–
O_2	0.09…0.15	0.10…0.15	–	–
N_2	< 0.006	< 0.006	–	–
Mechanical properties at 20 °C				
Elongation ratio δ, %, at 300 °C	28…40	28…40	37…50	17…26
Ultimate strength σ_B, kgf/mm² (MN/m^2)	22 (216)	22 (216)	15…19 (147…186)	22…34 (236…333)
Yield strength $\sigma_{0.2}$, kgf/mm² (MN/m^2)	11 (108)	11 (108)	12…16 (118…157)	20…30 (196…294)

For studies of the effect of iron on the properties of the Zr1%Nb alloy, samples of the alloy with an iron content from 0.012 to 0.192 wt.% were made at the NSC KIPT. Sponge (magnesium-thermal) zirconium was used as the base of the alloy, which provides an increase in the heat resistance of fuel element cladding in the conditions of the loss-of-coolant accident (LOCA) for light-water reactors.

Additional alloying of zirconium and its alloy Zr1%Nb with iron is promising in the development of alloys for reactors with high reliability and safety of operation, since an increase in the iron content in the zirconium alloy provides the material of the cladding tubes with the necessary creep resistance and hardening under irradiation, which ensures the design margin of stability and increases its corrosion and radiation resistance in the conditions of the nuclear reactor operation [5]. It is also known that with an increase in the iron content of the Zr1%Nb alloy in the structure of cladding tubes, the amount of Laves phase – $Zr(Nb,Fe)_2$ secretions increases, which favorably affects their corrosion resistance in water environment, especially for cladding tubes made of alloys based on sponge zirconium. Iron under the action of irradiation comes out from the Laves phase into the matrix with the formation of secondary finely dispersed precipitates and thus delays the formation of dislocation loops <c>-type which are responsible for accelerating the radiation growth of the alloy. At the same time it was established that as a result of additional alloying with iron the technological efficiency of the Zr1%Nb alloy decreases requiring the development of a new deformation and thermal scheme of tube manufacturing [6, 7].

Therefore, determination of the optimal iron content, which will not reduce the manufacturability of the alloy, but will increase the performance characteristics of the Zr1%Nb alloy and its service life, is an urgent task.

INITIAL DATA AND TASK SETTING

Thus, the problem statement can be formulated as follows:

You need to restore the *V (F, T)* dependency, where *V* is the corrosion rate, $mg/(dm^2 \cdot h)$; *T* is the observation period, h; *F* – iron content, %.

Features of this task:

– small sample of observations, which is associated with the high cost of conducting the experiment – (7 pairs of samples with a fixed mass content of iron for each pair were studied);

– lack of a priori information about the type of dependency you are looking for, which does not allow you to set its analytical model in advance.

To solve this problem, these features determine the feasibility of using the “machine learning” method.

The data of the results of experimental measurements of weight gain of Zr1%Nb samples after long-term corrosion tests are given in Table 2.

According to Table 2 formed a training sample (training dataset in the terminology of “machine learning”), presented in Table 3.

Table 2

Results of experimental corrosion tests of zirconium alloy samples with different iron content

Parameters	Iron content *F*, wt.%						
	0.012	0.042	0.072	0.102	0.132	0.162	0.192
1000 h testing							
Weight gain, mg/dm^2	36.3 35.3	36.6 33.3	32.8 31.8	28.7 29.9	31.3 31.5	33.3 32.4	32.54 33.98
Average weight gain, mg/dm^2	35.8	34.95	32.3	29.3	31.4	32.85	33.26
Average corrosion rate, mg/(dm^2·h)	0.0358	0.03495	0.0323	0.0293	0.0314	0.03285	0.03326
2000 h testing							
Weight gain, mg/dm^2	67.1 68.23	58 50.89	53.17 48.52	42 44.13	46.7 45.75	46.12 48.2	48.7 46.7
Average weight gain, mg/dm^2	67.665	54.445	50.845	43. 065	46.225	47.185	47.7
Average corrosion rate, mg/(dm^2·h)	0.033833	0.027223	0.025423	0.021533	0.023113	0.023593	0.02385
3000 h testing							
Weight gain, mg/dm^2	77.42 81.77	64.47 66.25	65.87 57.87	56.21 58.1	60.69 61.75	64.74 63.16	67.63 63.12
Average weight gain, mg/dm^2	79.595	65.36	61.87	57.155	61.22	63.95	65.375
Average corrosion rate, mg/(dm^2·h)	0.026532	0.021787	0.020623	0.019052	0.020407	0.021317	0.021792
4000 h testing							
Weight gain, mg/dm^2	92.74 92.42	85.18 88.75	80.79 85.24	73.2 79.1	81.03 82.58	83.19 85. 09	90.61 88.73
Average weight gain, mg/dm^2	92.58	86.965	83. 015	76.15	81.805	84.14	89.67
Average corrosion rate, mg/(dm^2·h)	0.023145	0.021741	0.020754	0.019038	0.020451	0.021035	0.022418

Table 3

Training Dataset

T, h	*F*, %						
	0.012	0.042	0.072	0.102	0.132	0.162	0.192
1000	0.0358	0.03495	0.0323	0.0293	0.0314	0.03285	0.03326
2000	0.033833	0.027223	0.025423	0.021533	0.023113	0.023593	0.02385
3000	0.026532	0.021787	0.020623	0.019052	0.020407	0.021317	0.021792
4000	0.023145	0.021741	0.020754	0.019038	0.020451	0.021035	0.022418

RESULTS AND DISCUSSION

Two-dimensional polynomial ridge regression was used to restore the dependence *V (F, T)*. The degree of the polynomial and the regularization coefficient were selected using a cross-validation procedure.

The dependency recovery algorithm is implemented in the Python programming language using the scikit-learn, pyswarm, and matplotlib libraries [8–10].

As a result of calculations, the degree of polynomial 3 and the regularization coefficient 0.6734 were selected.

The restored dependency is obtained in the following form:

$$V(F,T) = a_0 + a_1T + a_2F + a_3T^2 + a_4TF + a_5F^2 + a_6T^3 + a_7T^2F + a_8TF^2 + a_9F^3. \quad (1)$$

The coefficients of the approximating polynomial are shown in Table 4.

A three-dimensional visualization of the restored dependence of *V (F, T)* and the training dataset is shown in Fig. 1.

Table 4

Coefficients of the approximating polynomial

a_0	a_1	a_2	a_3	a_4
-0.68953395	-0.59437232	-0.22295343	0.36915152	0.0747594
a_5	a_6	a_7	a_8	a_9
0.33696609	-0.09272832	0.14789556	-0.05873025	-0.09225197

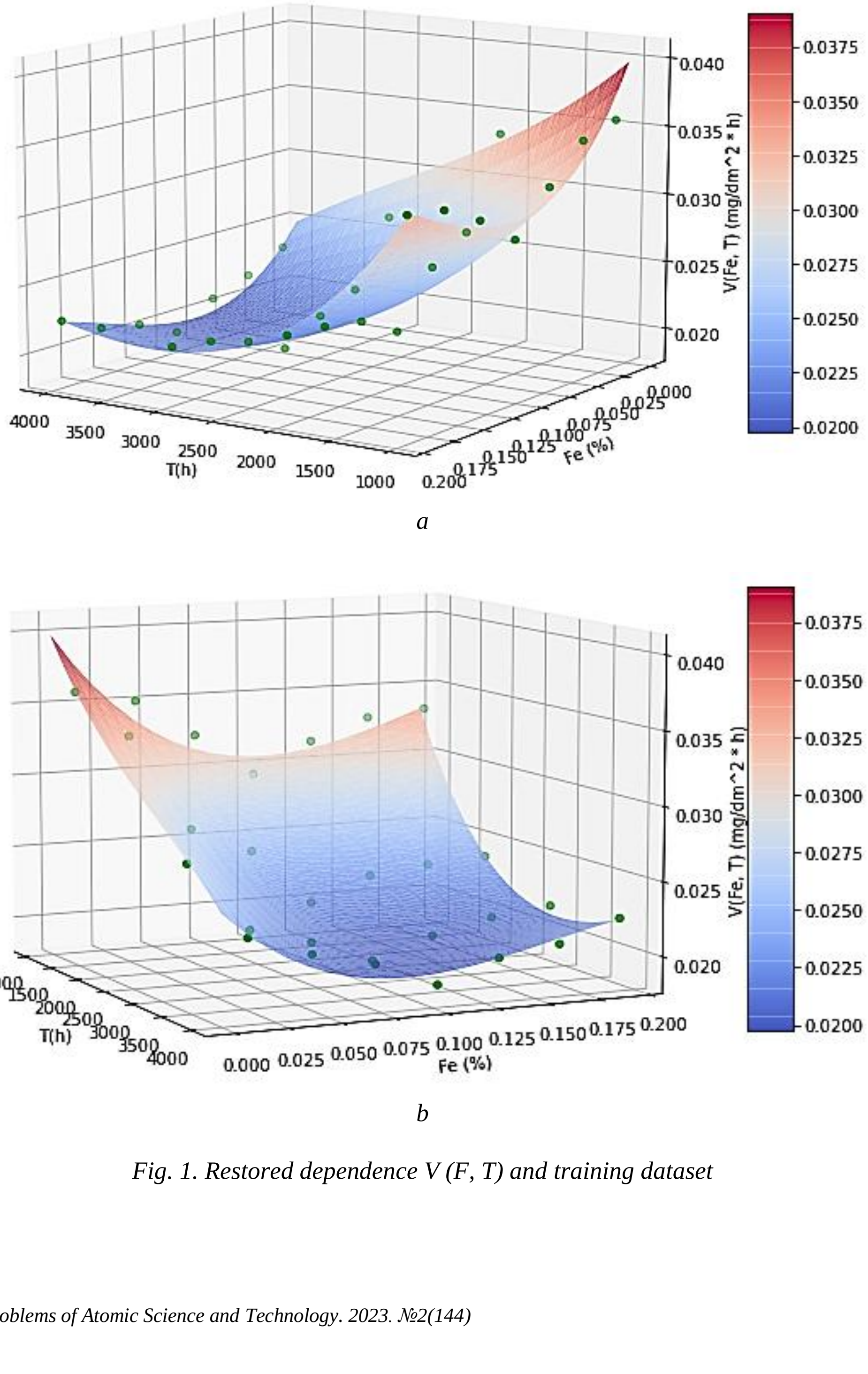
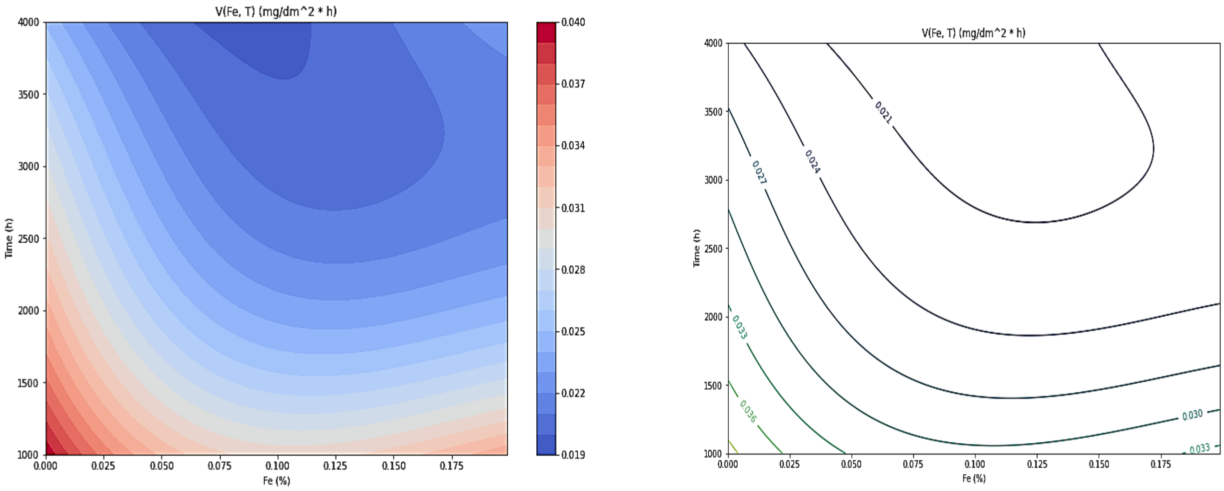


Fig. 1. Restored dependence V (F, T) and training dataset

The function *V (F, T)* is shown using contour graphs in Fig. 2.



a *b*

Fig. 2. Contour graphs of the restored dependency V (F, T)

In Fig. 3 shows sections of the approximating dependence *V* (*F*, *T*) for different values of the observation time.

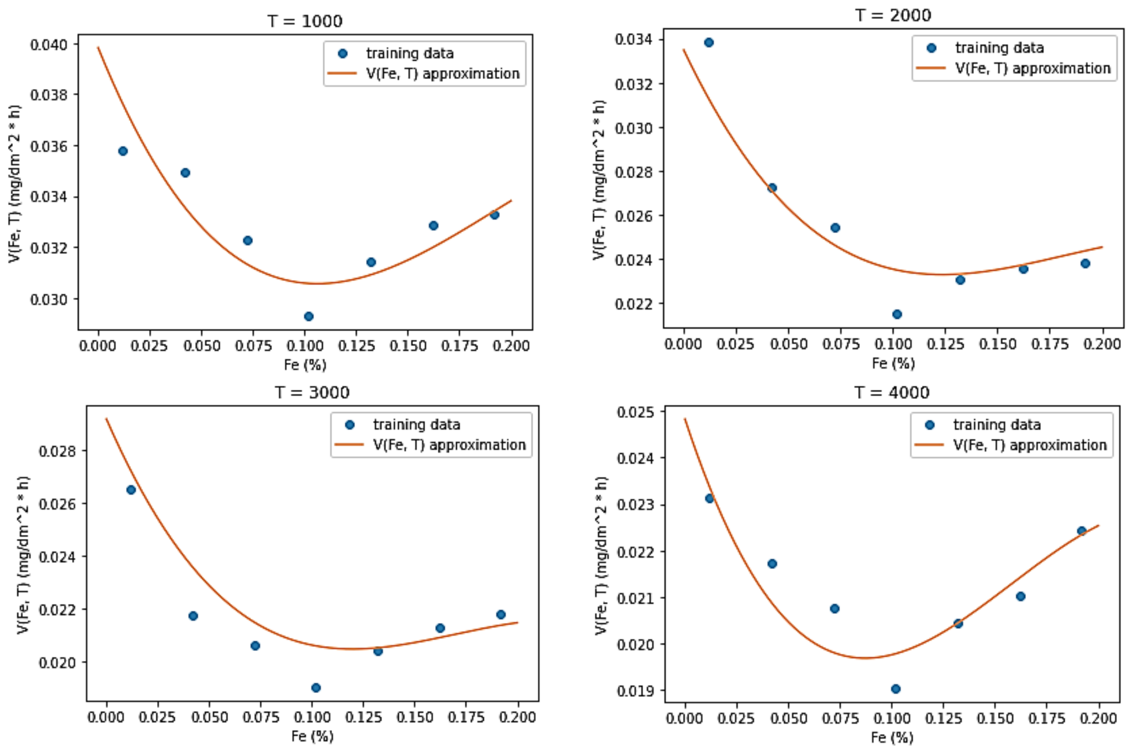


Fig. 3. Sections of the approximating dependence V (F, T)

CONCLUSIONS

Long-term corrosion tests of samples of alloy Zr1%Nb with different iron content, obtained by Ukrainian technology, in the water environment in composition and parameters (temperature 350 °C, pressure 16.5 MPa), which corresponds to the coolant of the primary circuit WWER-1000 when working at power, and mathematical processing of the results of these studies allowed us to determine the optimal amount of iron, which leads to an increase in the corrosion resistance of alloy Zr1%Nb in working conditions in the core of WWER-1000.

The results obtained allow us to conclude that the dependence *V (F, T)* at a fixed *T* has a pronounced one-extreme character, which indicates the presence of an optimal value of the mass content of iron, which provides a minimum rate of corrosion, and is localized around *F* = 0.1%. Consequently, the optimal amount of iron, which will increase the corrosion resistance of the alloy Zr-1%Nb under operating conditions in the core of WWER-1000, it is 0.1 wt.%.

The presence of an optimal value of the alloying iron content is typical for all zirconium alloys of Zr-Nb system. In the future, this opens up the possibility of producing fuel element cladding of increased corrosion resistance for the nuclear fuel cycle of Ukraine.

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МЕТОД ОПТИМІЗАЦІЇ ВМІСТУ ЗАЛІЗА В КОНСТРУКЦІЙНОМУ МАТЕРІАЛІ Zr1%Nb ДЛЯ ОБОЛОНОК ТЕПЛОВИДІЛЯЮЧИХ ЕЛЕМЕНТІВ ЯДЕРНИХ РЕАКТОРІВ АЕС

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Розглянуто математичний метод оптимізації кількості легуючого елемента заліза Fe в конструкційних цирконієвих сплавах Zr1%Nb оболонок твелів активних зон ядерних реакторів АЕС на основі фізичних експериментів для підвищення їх корозійної стійкості. Легування сплаву Zr1%Nb залізом Fe є перспективним при розробці технології виготовлення вітчизняних матеріалів оболонок твелів для реакторів з високою надійністю і безпекою. Для опрацювання результатів експериментальних досліджень утворення корозії сплавів цирконію з різним вмістом заліза було запропоновано математичний метод двовимірної поліноміальної гребеневої регресії з реалізацією його мовою програмування Python на основі теорії «машинного навчання». Застосування цього методу дозволило визначити оптимальне значення необхідної кількості легуючого елемента заліза (Fe) для цирконієвих сплавів Zr1%Nb оболонок твелів реакторів АЕС з водою під тиском.