UDC 539.2:536

INFLUENCE OF AN IRRADIATION ON ELASTIC MODULES OF METAL MATERIALS

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On the basis of the analysis of the literary data and the results received by authors, the general laws of influence of an irradiation in a wide interval of dozes and temperatures on elastic modules of metal materials are considered. Are analysed influence such as a crystal lattice type and a grain boundary role. Connection of change of elastic modules with temperature of tests, change of the parameter of a crystal lattice, radiation embrittlement of the reactor materials is investigated.

INTRODUCTION

Studying of influence of an irradiation on mechanical properties of materials continues to occupy one of leading places in radiation material science. Basically, processes *of plastic* deformation of the irradiated materials with various type of a crystal lattice in a wide interval of dozes and temperatures were investigated [1]. At the same time, many important questions of behaviour of the irradiated materials in *elastic area of deformations* remained not investigated. Among them - influence of an irradiation on elastic modules of materials.

Change of elastic modules at the big concentration of point defects for the first time has been theoretically investigated by J. Diens [2]. His calculations have shown, that elastic constants of copper should increase approximately for 10% on 1% interstitional atoms, but decrease, approximately, for 1% for 1 percent of vacancies.

However, results of other authors have shown difference of size of effect from Diens calculations not only on some orders (!) on size [3], but also and in a sign on the effect. For example, Zener [4] has predicted reduction of the shear modules, as vacancies, and interstitions. In Goland review [5] which have left more than through a quarter of a century after Diens work, it is marked «…a bad condition of the theory of influence of defects on elastic properties of materials …», and presence of ambiguity in prospective change under an irradiation of elastic constants.

The analysis of a modern condition of a problem shows, that despite of a long history, and great volume of the information on influence of an irradiation on elastic modules, the general laws of their change under an irradiation in a wide interval of dozes and temperatures are not established yet. Besides these changes till now are not taken into account properly in *models of evolution of defective structures*. Elastic modules enter into them as parameter, but change of the module is not taken into consideration. At the same time, it is necessary to expect "modular" effects at influence of an irradiation not only on change of mechanical properties (that is obvious), but also on structural – physical properties, such as swelling [6].

The purpose of work was an establishment of the general laws of influence of an irradiation on elastic modules of metal materials in a wide interval of dozes and temperatures of an irradiation.

1. MATERIALS AND A TECHNIQUE

The results of researches of influence of irradiation on elastic characteristics of materials with various type of crystal structure: V, Al alloys such as Д16АТ, austenitic steel Cr16Ni11Mo3, Ni, ferritic-perlitic steel Cr2NiMoV are presented.

The irradiation was carried out by various kinds of particles:

1. High-energy electrons with energy E = 225 MeV (Tirr ≤ 190 ^{0}C, dozes 0,002…0.2 dpa) on accelerator LU-2 GeV. Reliability of results of imitation reactor irradiation thus, is provided with a unique opportunity (e, γ) beams to create in a material not only point defects, but also complex defects, simultaneously saturate a material products of nuclear reactions [7].
2. Cr heavy ions with energy 1.8 MeV – on accelerator ESUVI up to dozes 1dpa at temperatures 200…290 ^{0}C.

Besides we had been used the experimental results of influence of an irradiation received earlier by neutrons in reactor Kiev Institute of Nuclear Researches up to a doze 1 dpa ($T_{irr.}$ = 350 ^{0}C).

For definition of elastic modules some methods have been used:

– "Static", based on measurement of a corner of an elastic site of a stress-strain curve (using Hook low);

– "Resonant", allowing to determine modules of elasticity, proceeding from frequencies of own fluctuations of samples [8];

– Using the nanoindentation method by a technique the Oliver-Pharr [9] was applied to definition of the module of elasticity of steel Cr2NiMoV, irradiated at temperature 290 ^{0}C, corresponding working temperature of reactor VVER-1000.

Understanding, that values *of elastic and shear module* of the various materials, determined by these methods, will differ, (that complicates their comparison among themselves, and also with the literary data), we considered their relative change under an irradiation. Thus, all changes have been correlated to parameter on which there is a comparison of a degree of damageability of all materials in radiating physics – to number of displacement on atom (dpa).

2. RESULTS OF RESEARCHES

2.1. INFLUENCE OF AN IRRADIATION ON ELASTIC MODULUS

On Fig. 1 stress-strain curve of the initial and irradiated vanadium are submitted. The irradiation, at very low dozes about thousand dpa, has resulted not only in increase in a yield strength, reduction of elongation, but also to increase in a corner of an inclination of an elastic site of a stress-strain curve related with elastic module of a material ($\alpha_2 > \alpha_1$). Simple estimations show, that relative growth of the module of elasticity thus makes about 5…6%.

In conditions of full absence of deformation hardening at the irradiated vanadium, when the amplitude of loading on a sample during deformation is not beyond elastic area, the role of elastic deformation after an irradiation grows. Besides change of a kind of a stress-strain curve unequivocally testifies to the main contribution to radiating hardening so-called «sources hardenings», connected with difficulty of generation of dislocations in irradiated, deformable at low ($T \leq 0.3T_m$) temperatures materials. The greatest effect from «sources hardenings» is observed on bcc-metals and alloys [1].

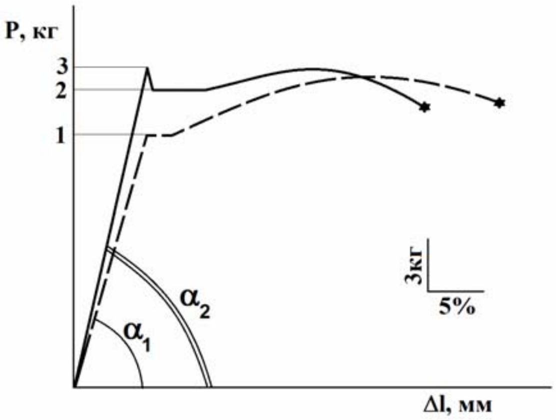

Fig. 1. Stress-strain curves initial (dotted line) and irradiated (a continuous line) up to $2 \cdot 10^{-3}$ dpa vanadium (20 °C). The point 1 –- corresponds to the loading, determining a yield strength of unirradiated vanadium; Points 2 and 3 correspond to the loadings determining, accordingly, the bottom and top yield strength of irradiated vanadium

On Fig. 2 results of influence of an irradiation on relative change of the module of elasticity of an aluminium alloy are submitted. As well as on samples of vanadium, the irradiation (Tirr = 100 °C) has led to to relative increase in the module of elasticity of alloy Д16АТ - ΔE/E on 5…7%. Apparently from figure, the size of effect poorly depends on temperature, i. e. carries athermal character.

Measurement of the shear module of reactor steel Cr16Ni11Mo3 has been carried out after reactor irradiation (Fig. 3). After a neutron fluence $2 \cdot 10^{25}$ cm^{-2} (about one dpa), the increase in the shear module in comparison with an initial condition made, on the average, 13…14%. It is important to note, that change of a corner of an inclination of temperature dependence of the shear module after an irradiation up to temperature of test 600 ^{0}C does not occur, that testifies about athermal character of effect of increase in the module as a result of an irradiation. In the same conditions of experiment influence reactor irradiations on the shear module of nickel has been investigated. In this case, the module has increased on 6…8%*.

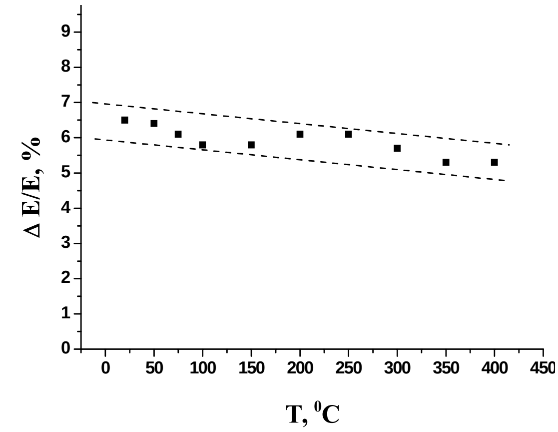

Fig. 2. Temperature dependence of change of the module of elasticity of alloy Д16АТ (0.11 dpa)

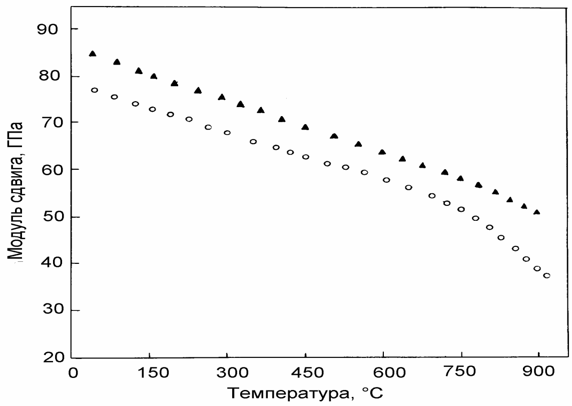

Fig. 3. Temperature dependence of the shear module of steel Cr16Ni11Mo3 initial (o) and irradiated in a reactor up to fluence $2 \cdot 10^{25} cm^{-2}$ (Δ)

2.2. COMPARISON OF EXPERIMENTAL RESULTS WITH THE LITERARY DATA

On Fig 4 change of elastic modules of materials with various type of crystal structure after an irradiation in reactors and on accelerators is submitted. Thus it was not done distinctions between the shear modulus and the module of elasticity, taking into account, that their changes are close on absolute size. There, where there were simultaneously data on the shear module and module of elasticity, the point on the schedule stands in the center of corresponding variability of data. On the gorizontal axis the doze of an irradiation in neutrons and dpa (for an irradiation on accelerators), on an axis of ordinates – relative change of modules *ΔM/M* (*Δ* G/G, *Δ* E/E) in percentage is postponed.

The submitted dependence can be broken conditionally into three areas on dozes and temperatures of an irradiation. The first – the basic area covering a wide interval of dozes (0.0001…1 dpa) and

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* Measurements were carried out by E.U. Grinik (Kiev Institute Nucl. Res. NAN of Ukraine).

temperatures of an irradiation (60…350 °C). The second area, there correspond dozes up to hundreds dpa, where at high temperatures of an irradiation passes swelling of materials. The third area corresponds to cryogenic temperatures of an irradiation, and dozes not exceeding 0.01 dpa.

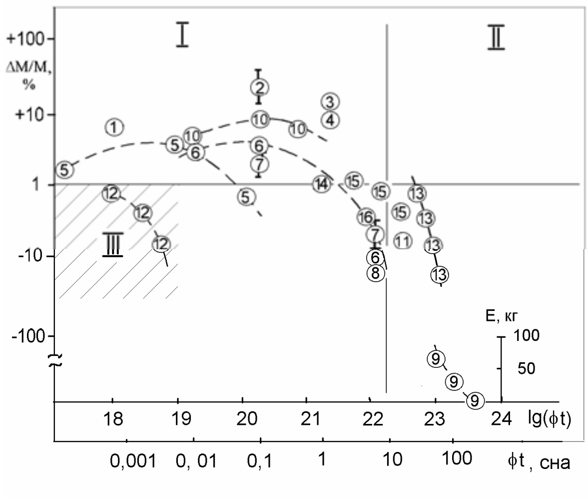

Fig. 4. Dependence of relative change of elastic modules (ΔM/M) under reactor and accelerator irradiation. 1 – vanadium, the present work; 2 – Al-alloy, the present work; 3 – steel Cr16Ni11Mo3, the present work; 4 – Ni, the present work; 5 – copper [10, 10a]; 6 – molybdenum [10,11]; 7 – tungsten [10,11]; 8 - ниобий [11]; 9 - steel 316 [12] For this material we have put an additional axis to show catastrophic falling the module of elasticity at swelling; 10 – iron [13]; 11 – ferritic - martensitic steel [14]; 12 – austenitic steel such as 03Cr20Ni16 [15]; 13 – austenitic steel Cr16Ni15Mo2 [16]; 14 – steel 15Cr2MoNiV, present work; 15 – austenitic steel 17Cr18Ni9 [17]; 16 – vanadium [33]

Area I. Analyzing the submitted data, it is possible to tell, that change of elastic modules submits to the same law: at low dozes of an irradiation (up to 10^{-1} dpa) rather small growth of modules which is replaced at big dozes not only their sharp decrease, but even by transition at dozes about 1dpa to negative values is observed.

Area II. This area corresponds not only dose – temperature interval of the radiation embrittlement, but also swelling effect. It is visible, that the irradiation leads to reduction elastic modules values already since dozes about several dpa [17]. As an example of extreme changes of the module, on Fig.4 the data on austenitic stainless steel of type 316 are submitted. Under the dozes about 180 dpa, elastic modules have fallen practically up to zero [12].

In the field of cryogenic temperatures (the area III, on Fig. 4) at all materials, irrespective of type of crystal structure is observed effect of decrease in modules under training [5]. At the same time, the doze dependences of change of modules in work have not been submitted. The search lead by us has shown, and in this interval of temperatures of an irradiation the same asymmetrical doze dependence curve with a maximum, as in 1 interval (Fig. 5) [18] is observed.

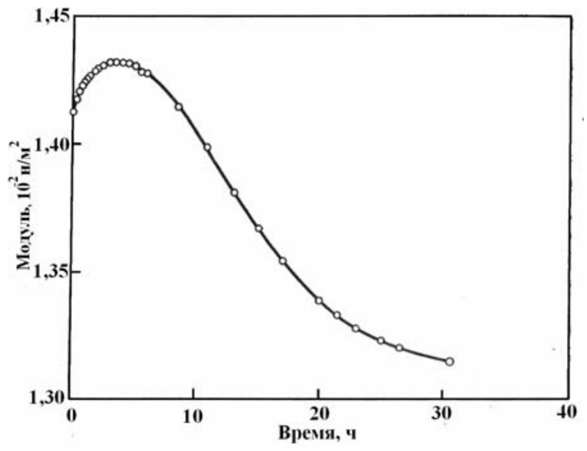

Fig. 5. Dependence of the module of elasticity of copper on duration of an irradiation alpha particles at 20 K [18]

2.3. RELATION OF ELASTIC MODULES CHANGE AND LATTICE PARAMETER OF THE IRRADIATED MATERIALS

Taking into account strong dependence of size of the elastic module (E) from parameter of a crystal lattice [19]:

$$E = Eo\ (a_o/a)^{\ m}, \qquad (1)$$

where E_o – the initial module of elasticity, a_o and a, accordingly, the initial and current parameter of a crystal lattice, an exponent m about 25.

For finding-out of the physical nature of effects of change of modules as a result of an irradiation, was of interest the analysis of change of lattice parameters of materials under an irradiation.

Results of the x-ray analysis of change of parameters of a lattice (Δa/a) molybdenum, iron, copper and the nickel irradiated in reactors on thermal and fast neutrons are resulted in [20-25]. The dependences received in works has nonmonotonic character and the form of an asymmetrical bell.

The results of the doze dependences change of parameter of a lattice of materials with various type of the crystal structure, irradiated with fast neutrons are presented on Fig. 6.

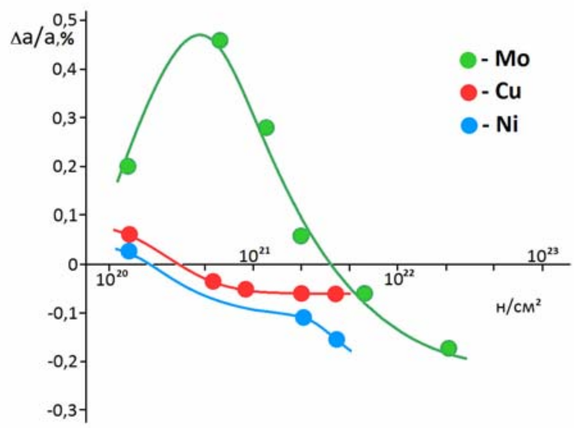

Fig. 6. Change of a lattice parameter of iron, molybdenum, and nickel under fast neutron irradiation. Mo u Ni-the given works [23, 25], Cu-works [24]

It is visible, that in comparison with bcc material (Mo), materials with fcc-type of a crystal lattice have shift of dependence in area of lower dozes, simultaneously to decrease in size of effect of an irradiation. This distinction finds the explanation within the framework of the experimental researches [26] which have established shift of a defect clusterisation threshold aside of lower dozes at fcc materials, in comparison with bcc, almost on two order of size, simultaneously with reduction in clusters concentration

3. DISCUSSION OF THE RECEIVED RESULTS

For an explanation of the effects described above, we use the representations advanced in works [22, 27] and becoming standard in physics of the radiation damages. In conformity with them, total change of parameter of a lattice of the irradiated material is determined *by a competition* of processes of interstitial atoms and vacancies (in various configurations) accumulation:

$$(\Delta a / a) = 1/3(C_i n_i - C_v n_v), \qquad (2)$$

where C_i, C_v – accordingly, interstitial and vacancy clusters concentration, and n_i and n_v an average of defects in them.

Let's consider the contribution interstitial atoms in doze evolution of Δa/a. According to works [28, 29], etc., it is necessary to pay attention to the following features:

– At the initial stages of an irradiation (since dozes about 0.001 dpa and till 0.05…0.1 dpa) the basic role in evolution of defective structure play interstitial atoms and their complexes.

– In a wide range of PKA energies from 0.5 up to 20 кeV interstitial atoms form almost exclusively single, di- and three-interstitials configurations. More than 99% of introductions in copper, and more than 99.5% of introductions in iron correspond to these configurations.

At low dozes of an irradiation of not exceeding 10^{-2} dpa linear dependence of parameter of a lattice on interstitial atom concentration n_i "works":

$$\Delta a/a = n_i \,\Delta\Omega / \Omega_o, \qquad (3)$$

where ΔΩ, Ωo – relaxation volume of interstitial atom in a lattice and atomic volume of the perfect crystal, accordingly.

It is possible to show, that with increase in a doze, with interstitial clusters growth, the size Δa/a becomes proportional $pm^{2/3}$, where p-density of clusters on m defects in everyone. Thus, growth of clusters is accompanied by reduction of concentration of point defects [30]. Thus, the contribution separate interstitial atom in the change of the lattice parameter, decreases with cluster growth. It should lead to a deviation from linear dependence Δa/a from a doze as it is submitted on a curve 1, Fig. 7.

As opposed to this, the contribution of single vacancies not only does not decrease, and even increases with growth of quantity of vacancies in clusters (see Fig. 7, a curve 2) [29]. It leads to increase in the negative contribution in (Δa/a) with increase in a doze, and in a result, - to the dependence submitted on Fig. 7, a curve 3.

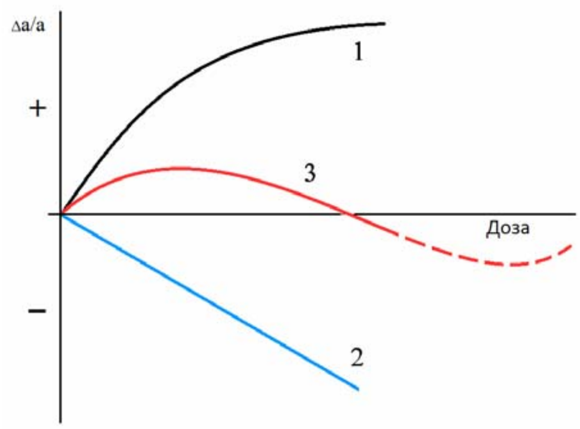

Fig. 7. Circuit of change of lattice parameter of the irradiated material in view of doze evolution features of the interstitial (a curve 1), vacancy (a curve 2) clusters. 3 – resulting curve. The dotted line designates transition to a stretching at the big dozes (area 2, Fig. 4)

The dotted part of a curve 3 on Fig. 7 reflects the forecast of change of lattice parameter at high dozes and temperatures of an irradiation. Authors believe, that growth on this part of a curve 3 corresponds to growth of porosity in the irradiated materials at dozes about tens dpa when in a material the condition of a hydrostatic stretching of a lattice is formed.

This simple model allows on the doze dependencees data of lattice parameter of materials to predict behaviour of their modules.

The irradiation can change elastic modules several ways:

– To increase, owing to strengthening of atomic bonds in a lattice at introduction in it interstitial atom, on what specified still Diens [2];

– To reduce, owing to easing of atomic bonds in the crystal lattice, caused by vacancies and them clusters;

– To increase, owing to influence of radiation defects on fastening of a dislocation [5].

So, at introduction in a crystal interstitial atoms, and growth thus of lattice parameter there can be an additional interaction on distances, smaller, than distance between the nearest atoms that eliminates the seeming contradiction with the formula (1).

The increase of the vacancy clusterization with doze will lead to decrease of the modules due to easing interatomic forces. One of proofs of its, are results of measurement of modules on the quenched samples. As a rule, the quenched samples have the lowest values of the shear module in all an interval of temperatures [32].

And, at last, the direct proof of a main role of the vacancy clusterization in falling elastic modules are the given works [12] testifying to falling up to zero of the module of elasticity at a swelling level ~ of 30% and zero plasticity.

During growth of the elastic module in the given conditions of an irradiation (dozes – there is less than one dpa, the temperature of an irradiation is lower $0.35\,T_m$) *the effect of amplification of the covalency component of the interatomic bonds in the irradiated metals* [31] established by us earlier can play essentially important role.

Influence of test temperature and grain boundaries effects. The results submitted on Fig. 2, 3 (fcc - materials), and also of some materials with bcc - type of a crystal lattice [33] show, that in a wide interval of temperatures of tests (up to 0.45…0.5T_m) change of modules as a result of an irradiation, practically does not depend on temperature.

Same athermal dependence is observed and for radiation hardening the materials irradiated in reactors, high-energy electrons with energy E = 225 MeV, heavy ions, that is, in conditions of the irradiation creating of the cascades of displacement of atoms [34].

At the same time, at some temperature (about 750 ^{0}C for austenitic steel) on temperature dependence of the shear module there is an excess (bump), the most strongly pronounced at unirradiated material (see Fig. 3). This excess is connected to temperature of the grain boundary relaxations beginning, and determined by intensity of the grain boundary deformations processes [8].

The analysis, the researches executed in this direction in Kiev Institute of Nuclear Researches under V.S. Karasev and E.U. Grinik's management has shown, that *is direct* under action of reactor irradiations, maxima of the grain boundary relaxations are shifted in area of low temperatures, and shear modules of iron and nickel decrease (Fig. 8) [35].

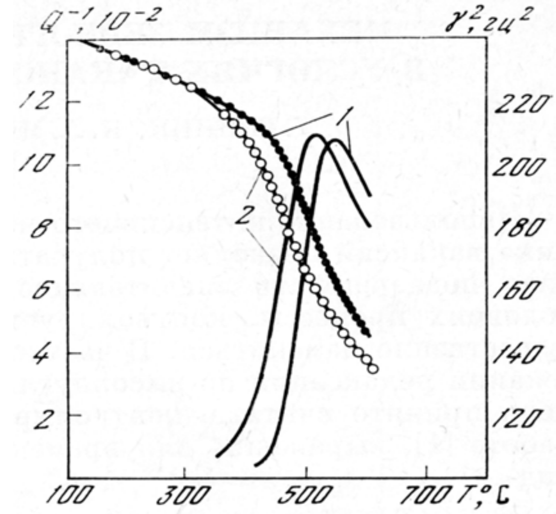

Fig. 8. Temperature dependence of internal friction and the shear moduls of iron in an initial condition (1) and during an irradiation neutrons (2) [35]

The calculations which have been carried out in work [35] have shown, that change of the grain boundary internal friction temperature maximum and values of the module can be connected with vacancy supersaturation, achieving values about 6, and her influence on the grain boundary relaxations processes and grain boundary sliding. The same authors, at an irradiation of alloy Fe-B^{11} [36] had been established neutron flux effect, consisting that values of the module during a neutron irradiation are lower, than at the stopped reactor (in an interval of temperatures of test 350…630 ^{0}C). Displacement aside low temperatures the grain boundary peak of internal friction and reduction of the module also has been fixed in the work [37] executed on samples of nickel, irradiated high-energy electrons with energy E = 225 MeV.

As the answer to a question on concrete mechanisms of influence of an irradiation, on grain boundary properties, we shall refer to results of work [34], where it has been shown, that as a result of an irradiation, grains boundaries become not saturation sinks for so-called *brought grain boundaries dislocations,* that provides increase of intensity of grain boundary deformation process in the irradiated materials. In result, shift grain boundary maximum in area of low temperatures, and decrease in the module corresponding to it (see Fig. 8) takes place.

Radiation embrittlement and change of elastic modules. It is necessary to note, that the dependence submitted on Fig. 4 reflects also behaviour of modules in various intervals radiation embrittlement. The stage of growth corresponds (in most cases) to an interval of temperatures which can be named «low temperature radiation embrittlement», and a stage of falling - middle, - and high-temperature radiation embrittlement (from above 350…500 ^{0}C).

At the description of these effects it is necessary to take into account the modern sinergetic approach to process of deformation. One of the basic concepts of this approach is the concept about hierarchy micro-, meso- and macro-scales of structural levels of plastic deformation. According to the theory of processes of self-organizing of structure in the deformed material, characteristic scale of areas in which energy dissipates at deformation, $L_s \sim G$, where G – the shear module [38]. In fcc – and the bcc- materials irradiated up to dozes, not exceeding a some dpa and temperatures till 0.35 T_m (area 1, Fig. 4), elastic modules grow and, according to the mentioned above theory, should increase and scale of L_S-areas of self-organizing of the disspative structures in the deformed irradiated material, as has been established in work [34].

In the field of 2, at high dozes (180 dpa) and temperatures of an irradiation (400…500 ^{0}C), at values of swelling more than 30 %, in conditions of absence of shearing modes of plasticity, hydrostatic tensile stresses result to sharp decrease in the elastic module and to full embrittlement of steel already at stresses became much lower than a yield strength of a material [12].

CONCLUSIONS

The analysis of results of research of influence of an irradiation high-energy electrons, heavy ions, neutrons, has allowed to establish, for the first time, doze dependence of change *of elastic modules* of materials with various type of a crystal lattice in a wide interval of dozes and temperatures of an irradiation which have identical, so-called "bell-like" character.

- At low dozes (less than 10^{-1} dpa) rather small growth of modules which then is replaced by their reduction is observed. Thus, change of modules remains positive down to dozes corresponding a some dpa.

- Correlation of changes of elastic modules and parameter of a crystal lattice of metals with a doze is established. It is shown, that such dependences correspond to a competition of processes interstitial and vacancy clusterization.

- Change of modules has athermal character down to the temperatures corresponding to the beginning of processes of the grain boundary relaxation.

- Change of the shear module under irradiation is according to laws of development of the radiation emrittlement of materials, testifying about growth as a result of an irradiation, of scale of a structural level of deformation of a material.

Authors thank V. Savchenko, I. Kolodija, M. Tihonovskogo for the help in carrying out of experiments, E. Krajnjuka for the help by preparation for the publication.

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Article received 05.03.2014

О ВЛИЯНИИ ОБЛУЧЕНИЯ НА УПРУГИЕ МОДУЛИ МЕТАЛЛИЧЕСКИХ МАТЕРИАЛОВ

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На основе анализа литературных данных и результатов, полученных авторами, рассмотрены общие закономерности влияния облучения в широком интервале доз и температур на упругие модули металлических материалов. Проанализированы влияние типа кристаллической решетки и роль границ зерен. Изучена связь изменения упругих модулей с температурой испытаний, изменением параметра кристаллической решетки и радиационным охрупчиванием реакторных материалов.

ПРО ВПЛИВ ОПРОМІНЕННЯ НА ПРУЖНІ МОДУЛІ МЕТАЛЕВИХ МАТЕРІАЛІВ

І.М. Неклюдов, В.М. Воєводін, І.М. Лаптев, О.О. Пархоменко

На основі аналізу літературних даних та результатів, отриманих авторами, розглянуто загальні закономірності впливу опромінення в широкому інтервалі доз та температур на пружні модулі металевих матеріалів. Проаналізовано вплив типу кристалічної гратки та роль границь зерен. Вивчено зв'язок зміни пружних модулів із температурою випробувань, зміною параметра кристалічної гратки і радіаційною крихкістю реакторних матеріалів.