Results of Monitoring, Using High-Resolution Neutron Diffraction, of Radiation-Induced Damages in Claddings of Fuel Pins After Their Performance in the Reactor BN-600 as a Ground for Prolongation of Their Life Expectancy

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Abstract. Fast neutron irradiation gives rise to rather complex processes developing in the fuel element claddings that lower their technical characteristics and restrict time of safe exploitation. Neutron diffraction studies open up a possibility to monitor them even at a stage of incubation period and thereby to promote development of reliable methods for life expectancy prolongation of the reactor components. An important advantage of these methods is minimal manipulations needed for work with high radioactive samples.

Key Words: fast reactors, radiation-induced damages, austenitic steels, neutron diffraction.

1. Introduction

Evolution of properties of any functional material in the process of its exploitation is governed by changes in its microstructure; namely, formation and evolution of radiation defects, micro precipitations, changes in the phase content, texture, grain size, and state of grain boundaries, etc. Understanding of fundamental mechanisms of these phenomena is of great importance for both improvement of material properties and development of methods of prolongation of life expectancy of the related articles.

This in full measure concerns cladding materials of the fuel pins. However, investigation of changes arising in these claddings is rather a difficult task because of high induced radioactivity of these materials. As a result, for the performance of such works, special equipment is required, as well as advanced system of preparation and transportation of the samples and, first of all, highly qualified personal.

In Zarechny-city, where the Beloyarsk Nuclear Power Plant (BNPP) is situated, there exists a unique combination of such facilities. First, in the nearest neighborhood with the BNPP, the Institute of Nuclear Materials (INM) is situated which hosts, along with the research reactor IVV-2M, equipment for studies of samples from the fuel elements after their exploitation in the BN-600 reactor, in particular, of their physic-mechanical properties, microstructure, and corrosion. Second, the Institute of Metal Physics (IMP) build up on the INM premises the Neutron Material Science Complex (NMSC) for works in the field of radiation physics and solid-state physics with the use of neutron diffraction methods. At present, the NMSC is the only center in Russia where neutron diffraction studies of highly radioactive materials are fulfilled.

Structure investigations with the use of neutron diffraction present important advantages. Unlike X-ray experiments, owing to large neutron-penetration depth, massive samples and industrial products can be explored. This method is rather sensitive to the content and distribution of light elements, such as carbon or nitrogen, and alloying metals (titanium, manganese, etc.), which essentially affect steel properties. With the help of neutron diffraction it is possible to measure both the average-over-volume lattice deformation and stresses within grains and, at the same time, to determine the crystallographic anisotropy of deformations and, the more, in case of multiphase material, stresses for each phase proper. Moreover, the texture of material, average size of coherently scattering regions, and phase composition can be examined simultaneously. Besides, this method demands minimal time for manipulation with radioactive samples, giving the possibility to operate with high radioactive materials. The experience of long-term cooperation of BNPP, INP, and IMP RAS and combination of their experimental facilities allow one to provide systematic complex studies of the radiation-induced effects in the materials of the fuel pin claddings of the BNPP fast reactors.

Up to now, we have gained sufficient experience of such studies. In particular, for last decade, we have been studying systematically the effects of irradiation with fast neutrons on different austenitic steels, such as EK164 and ChS68, which were used for production of the fuel pins for the BN600 reactor.

2. Preliminary studies

The main methods of improvement of radiation resistivity of reactor steels are doping with different elements and thermo-mechanical treatments resulting in the formation of systems of traps where recombination of the point defects takes place [1]-[2]. In principle, understanding of such systems and mechanisms of their functioning is a key to creation of radiation-induced materials.

As the elemental content of the steels used in fast reactors is very complex, to understand the radiation-induced processes proceeding in them and to develop special methods for their observance, we carried out experiments with a set of FCC model systems. They were pure nickel and nickel doped with different elements, ferro-nickel invar, and also ageing austenitic steels with different (carbide and intermetallic) types of ageing [3]-[5].

For example, we have managed to record two competitive processes in these aging steels arising under irradiation of the materials by fast neutrons. The first one is the radiation-stimulated relaxation of the microstresses arising in the material because of precipitation of carbide or intermetallic phases in the process of artificial ageing preceding the irradiation. The other alternative process is generation of radiation damages in the form of vacancy clusters and dislocation loops, which causes growth of the lattice micro deformations. It is important to note that the temperature of irradiation in these experiments (in the IVV-2M reactor core) made up about 60°C, which provides low thermal mobility of the substitutional atoms. In the case of large initial internal stresses and small fluence, the first process prevails. In the case of large fluence, the process of radiation-induced defects generation dominates resulting in the appearance of additional microstresses in the samples. Note that in the course of both aging and irradiation with fast neutrons evolution of microstresses depends on the type of precipitating particles and their concentration.

3. BN600 reactor steels

In collaboration with colleagues from INM we carried out neutron-diffraction studies of the microstructure of samples of the fuel pin claddings of the BN600 reactor produced from the austenitic steel EK164. Neutron diffraction patterns were taken with a diffractometer D7a at the IVV-2M reactor. Experimental data were analyzed with the program Fullprof [6] using of the tool resolve function.

First of all, it has been clarified that our analysis allows easily detecting even small variations in the initial state (prior to irradiation) of the samples produced from EK164 but belonging to different sample lots. In Table 1 one can see the values of the texture coefficients of the fuel pin claddings from three lots. The diffraction patterns of all samples showed practically the single-phase FCC-structure with very close values of lattice parameter a.

The texture coefficients were determined in accordance with the technique proposed in [7] by formula:

$$TC_{h_ik_il_i} = \frac{I(h_ik_il_i)}{I_0(h_ik_il_i)} \Big/ \frac{1}{n}\sum_{j=1}^{n} \frac{I(h_jk_jl_j)}{I_0(h_jk_jl_j)} \qquad (1)$$

where $I(h_jk_jl_j)$ are the intensities of n peaks included in our analysis and I_0 are the intensities of the powder reference.

Since in experiments the samples were pieces of the cold rolled tubes standing vertically (i.e. neutrons were reflected from the planes parallel to the tube axis), then the value of the texture coefficient allows a conclusion to be made about the preferable texture of the corresponding grain planes along the tube axis. As is seen from Table 1, the maximal values of the texture coefficient are typical of the (220) planes.

TABLE 1: LATTICE PARAMETERS, TEXTURE COEFFICIENTS, MICRO DEFORMATION ALONG THE CRYSTALLOGRAPHIC DIRECTIONS AND DISLOCATION PARAMETERS FOR NON-IRRADIATED SAMPLES OF STEEL EK164.

Sample Number	**1**	**2**	**3**
a, Å	3.5844	3.5854	3.5842
TC_{111}	0.28	0.39	0.45
TC_{200}	1.36	0.97	0.87
TC_{220}	2.22	2.45	2.39
TC_{311}	0.58	0.89	0.89
TC_{222}	0.22	0.30	0.39
TC_{400}	1.35	0.99	1.00
$\Delta d_{111}/d_{111}$, 10^{-4}	29.1	27.0	25.7
$\Delta d_{200}/d_{200}$, 10^{-4}	41.9	37.7	33.3
$\Delta d_{220}/d_{220}$, 10^{-4}	32.8	30.1	27.8
$\Delta d_{311}/d_{311}$, 10^{-4}	36.4	33.1	30.0
$\langle \Delta d/d \rangle$, 10^{-4}	35.2	32.1	29.3
ρ, 10^{10} cm^{-2}	1.14	0.88	0.71
C_S	0.64	0.62	0.42

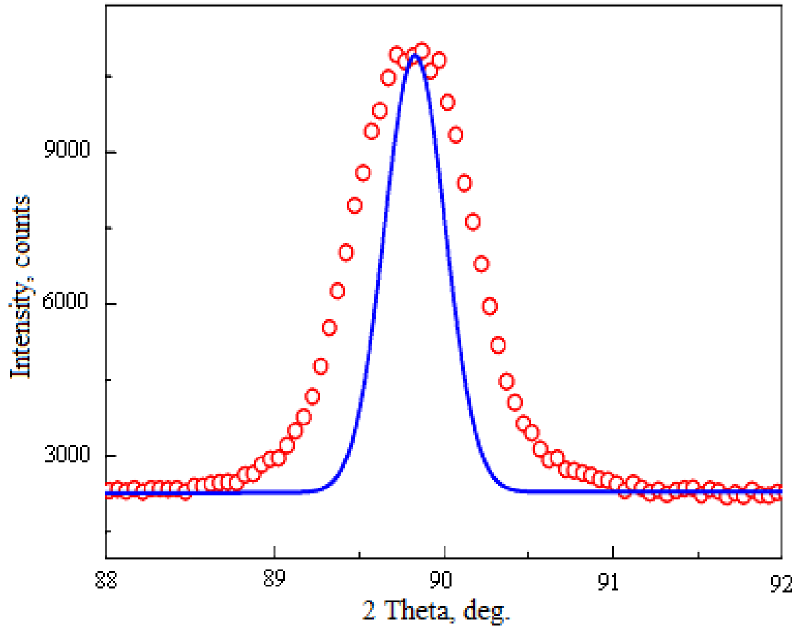

FIG.1. Experimental (circles) and reference (line) diffraction peak (311) for sample No 1.

The second specific feature of the experimental neutron diffraction patterns of the samples under study is the peak broadening, as well as a change in the form of diffraction peaks with respect to the reference (instrumental) peaks (FIG.1).

Deviations from the reference line and the peak broadening can be connected with either small sizes of coherent-scattering regions or development of microstresses within the material bulk. We treated the neutron-diffraction patterns by the Rietveld method, taking into account the anisotropy broadening. It was found that the size of coherent-scattering regions (CSR) did not decrease. The broadening was caused only by internal microstresses. Table 1 shows also the results of our calculations of the microdistortions along different crystallographic directions and their average volumes.

It should be noted that the experimental neutron diffraction patterns show a significant difference of the reflection broadening in different crystallographic directions resulting in a clear difference of micro deformations in these directions (Table 1). This effect is probably caused by changes in the dislocation structure. We evaluated the density of dislocations using the modified method of Williamson-Hall [8 - 11]. The samples of non-irradiated claddings are found to be characterized by scattering values of the dislocation density ρ and relative fraction of the screw and edge dislocations C_S (Table 1).

Irradiated samples for our studies were cut from different parts of the fuel pins at different height in the BN600 core, i.e. their exploitation conditions were characterized by different temperature and fast-neutron flux densities. For example, in Table 2 one can see the data collected for the samples cut from the same fuel pin with the indication of their coordinate Z (vertical distance from the bottom of active zone, height of which was 1030 mm), temperature of irradiation, and the neutron dose.

FIG. 2 shows the neutron diffraction patterns for the samples prepared from the fuel pins in the initial state and after their work in BN600 reactor.

TABLE 2: EXPLOITATION CONDITIONS FOR THE STUDIED SAMPLES MADE OF STEEL EK164 AND MICRO DEFORMATIONS ALONG THE CRYSTALLOGRAPHIC DIRECTIONS

Z, mm	**Doze, dpa**	**T_{irr}, °C**	**$\Delta d_{hkl}/d_{hkl}$, 10^{-4}**			
			(111)	**(200)**	**(220)**	**(311)**
908	49.1	628	20,21	29,81	22,99	25,73
474	72,3	528	29,99	50,06	36,07	41,82
225	70.8	463	27,83	49,81	34,66	40,95
54	48.0	419	26,28	53,27	35,03	42,73
12	32,8	407	32,38	53,16	38,65	44,6
-118	14,5	389	33,07	59,01	41,12	48,55
-734	0.6	370	21,22	35,08	25,41	29,38

As it is seen from FIG. 2, the qualitative appearance of the patterns did not change (including peak intensities ratio), in spite of rather high doses of fast neutrons. The material preserved its FCC structure successfully. However, at the same time, the width and form of the peaks changed significantly. The line form can be characterized via describing the experimental form with the use of the so-called Pseudo-Voigt function (linear combination of Lorenz and Gauss functions [6]).

In FIG. 3, one can see the dose dependences of the peak width and coefficient η (relative fraction of Lorenzian in Pseudo-Voigt)

It is seen that the maximal broadening of reflections is observed at relatively low temperatures of irradiation, lower than 400 ^{O}C, and doses less than 15 dpa. These results agree well with the concepts on the formation under these conditions of a large amount of interstitial dislocation loops and small vacancy voids and on a poor relaxation ability connected with the low mobility of substitutional atoms at these temperatures.

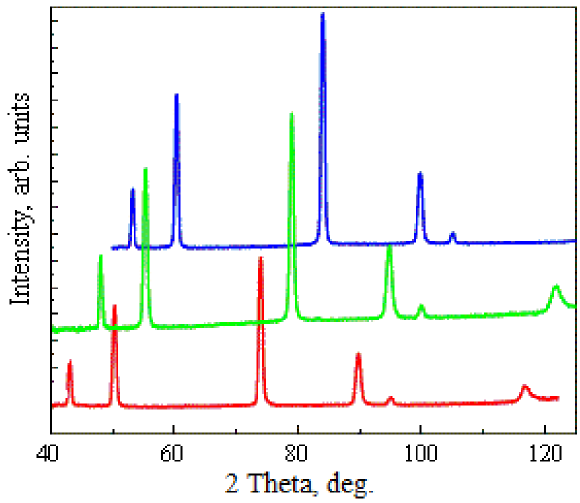

FIG. 2. Experimental neutron diffraction patterns for the samples prepared from the BN600 fuel pins: blue – initial state; green – the doze of 14.5 dpa at T=389 ^{O}C; red – the doze of 72.3 at T=528 ^{O}C. For better illustration purposes, the patterns are shifted by the zero angle and height.

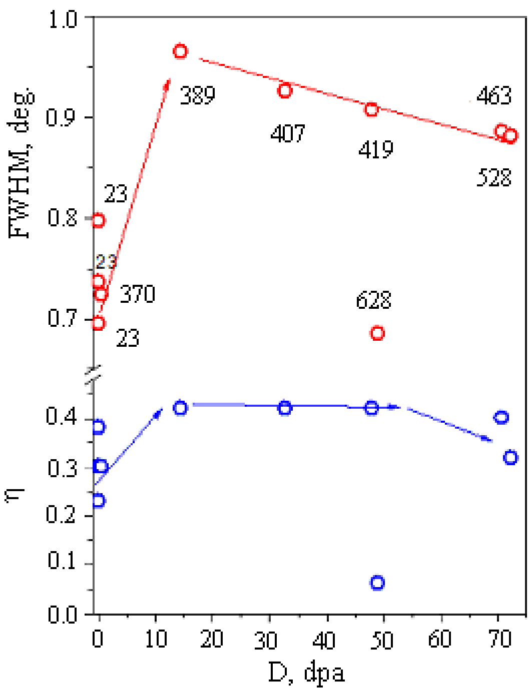

FIG.3. Dose dependences of the (311) peak width and coefficient η (relative fraction of Lorenzian in its form. Temperatures of irradiation are given in the panel near the circles.Temperature 23 C corresponds to not irradiated samples.

Yet, with increasing the dose of fast neutrons, the plot shows narrowing of the reflection, which could be traceable to the fact that the doses were collected in the core regions under enhancing irradiation temperatures. For EK164 steel, no pronounced correlation between the lattice parameter of the material and the irradiation dose was observed up to the dose of 75 dpa.

The neutron diffraction results on the lattice microdeformations in different crystallographic directions look more interesting. FIG. 4 shows the microdeformation values <Δd/d>, averaged over all directions (qualitatively, the plots for individual directions have the same form).

It is seen that, with elevating the dose of fast neutrons, at a relatively low temperature of irradiation (T<400 ^{O}C) the microdeformations increase rapidly. However, at temperatures above 400 ^{O}C there is observed a decrease in the microdeformation values even when the damaging dose grows. The role of irradiation temperature can be distinctly observed at the damaging dose of 48 – 49 dpa (FIG.4): with increasing the temperature from 419 ^{O}C to 628 ^{O}C, the values of <Δd/d> lower from 40 to 25, i.e., by a factor of 1.6. Hence, one can judge the competing processes that take place upon irradiation of the material with fast neutrons and heating. At low temperatures, the impurity atoms are unable to intensively leave the solid solution. The formation of interstitial dislocation loops results in the growing microstresses. The enhancement of temperature intensifies the processes of migration of both intersititial impurity atoms and substitutional atoms over vacancies, whose concentration is much higher than the thermally equilibrium one, which causes the stress concentrators to go out of the lattice and secondary phases to form. As a result, the microstresses in the lattice diminish, which is registered in the neutron diffraction experiments. Note that the dose dependence calculated from the neutron diffraction data on the dislocation density demonstrates the same behavior as in FIG. 4.

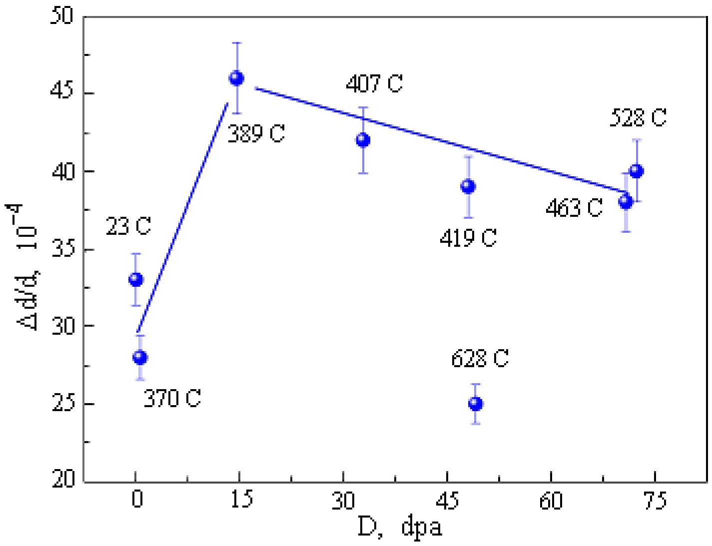

FIG. 4. Dependence of microdeformations averaged over directions for the samples of steel EK164 on irradiation dose. Temperatures of irradiation are given in the panel.

The working conditions of a part of the fuel pin (temperature and density of the fast neutron flux) are controlled by its position in the core. Consequently, to monitor the performance the fuel pins, a direct analysis of correlations between our results and coordinates of the samples studied, which are shown in Table 2, is reasonable. FIG. 5 presents the obtained dependence of the dislocation density on the position of samples in the reactor core.

The accumulation of defects in a material is known [12] to be accompanied by changes in the width and form of reflections, as well as emergence of wide diffuse maxima near the reflections of the main and precipitated phases. Moreover, because of the microstresses caused by the presence of defects, broad wings appear near the reflections, which manifest themselves in the change of the reflection form from the Gaussian to Lorenzian one, in agreement with our experimental results (FIG.3). As an example, the dependence of the coefficient of the line form η on the position of the sample in the core is shown in FIG.6.

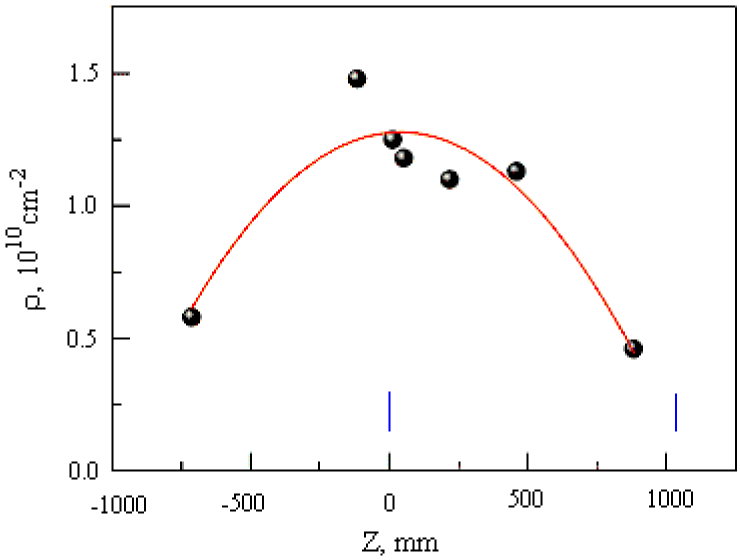

FIG. 5. Dislocation density as a function of the relative localization of samples in the reactor core (shown by blue vertical lines)

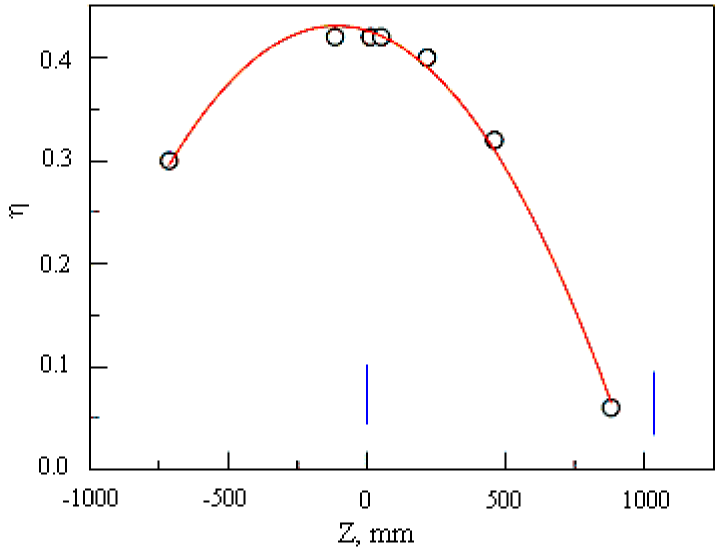

FIG. 6. Dependence of the coefficient of the reflection form η on the position of the sample in the core (designated by blue vertical lines)

The main problem of using of the fuel pin claddings from austenitic steels is the tendency of the material toward vacancy swelling. Interestingly, the maximal swelling of the fuel pins in the BN600 reactor is observed in the region with the coordinate Z of approximately (200 – 510) mm at a height of the core of 1030 mm, which depends on the parameters of the cladding-fabrication process and distribution of doses and temperatures of irradiation. The maximal dislocation density determined from the neutron diffraction measurements, which is largely contributed by the formation of interstitial dislocation loops, matches just this range. Similarly to the edge dislocations in the FCC lattices, dislocation loops are preferable sinks for interstitials and play a harmful role favoring the predominance of the saturated concentration of vacancies over interstitials, thus resulting in enhancement of swelling.

FIG.7 shows, as an example, the electron microscopy data taken from the samples made from different parts of the fuel pins produced from austenitic steel ChS68 close in properties to steel EK164.

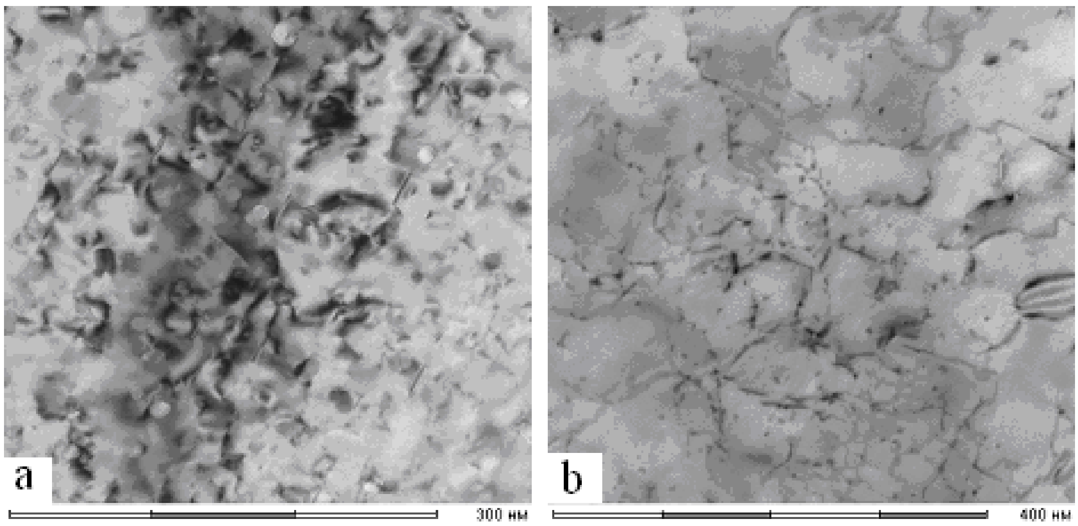

FIG. 7. Dislocation structure of the fuel pin cladding.
a - Core bottom, Tirr=415 ℃, D=50 dpa; b – Core top, Tirr=590 ℃, D=65 dpa.

The accumulation of dislocations demonstrated by FIG.7 is supported by our neutron diffraction data.

4. Conclusion

The plots shown in FIGs. 1- 6 confirm the opportunity of applying neutron diffraction in combination with other methods in monitoring of the material state for the fuel pin cladding. Besides, they show up certain latent changes that are precursors of the structure changes followed by the changes in the physico-mechanical properties.

With the use of neutron diffraction methods, irradiation-induced phenomena in austenitic steels, in particular, in materials for the fuel pin claddings, can be traceable. For example, they are precipitation and dissolution of particles, changes in the grain parameters, and behavior of internal microstresses in the system. It is established that though up to quite high doses employed, the FCC structure of steel EK164 is retained, the microstructure of the material is essentially dependent on the dose and temperature of irradiation. Neutron diffraction studies favor understanding of the phenomena that develop in the reactor materials in the process of their exploitation and can be very fruitful for both designing of advanced materials and determination of the life expectancy of the existing articles.

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