

Investigation of Radiation-Induced Swelling of EK-164 Steel, an Advanced Material for BN-600 and BN-800 Claddings

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Abstract. One of the main issues on fast reactors is to improve their economic efficiency. Nowadays austenitic stainless steel ChS-68 is used as a standard material for fuel pin claddings of BN-600 reactor ensuring damage doses up to 87 dpa. Post irradiation examination results show the residual life of fuel pins with possible maximum damage dose up to 92-95 dpa, corresponding to fuel burn-up to 12-13 % FIMA. At VNIINM a prospective austenitic stainless steel EK-164 (16Cr-19Ni-2Mo-Ti-Si-V-P-B), more resistant to radiation-induced swelling than ChS-68 steel, has been developed to extend service life of fast reactor fuel assemblies.

A trial operation of ChS-68 and EK-164 combined fuel assemblies has been carried out to ensure the operating capacity and assess the capability to improve operational characteristics of fuel pins with EK-164 claddings. At the initial stage maximum damage dose during operation of two test fuel assemblies is 74 and 77 dpa, respectively. Post irradiation examination confirms the advantage of EK-164 steel in terms of radiation-induced swelling resistance.

Manufacturing technology of claddings used as a material for fuel pins operated to maximum damage dose in the range between 84 and 96 dpa has been improved, considering investigation results for structure and properties of EK-164 claddings. Post irradiation examination shows that fuel pins retain their operating capacity and have sufficient residual life. Therefore it is possible to predict the operation resource at damage doses over 110 dpa.

The paper aims to investigate radiation-induced swelling of EK-164 claddings at different temperatures and neutron irradiation doses, and to distinguish radiation-induced porosity characteristics at different irradiation temperatures as well.

Key Words: austenitic steel, neutron irradiation, radiation-induced swelling.

1. Introduction

Radiation-induced swelling is one of factors limiting service life of structural materials in fast reactors. It significantly affects reactor service life.

The paper aims to investigate radiation-induced swelling of EK-164 claddings at different irradiation temperatures and doses, and identify radiation porosity dependence on irradiation temperature in particular.

2. Materials and testing procedure

Specimens of two cladding lots made of cold-worked EK-164, induction arc remelting (in what follows EK-164), have been examined. The steel composition is given in [1].

Cladding from the first lot were manufactured at Joint Stock Company "Pervouralsk New Pipe Plant" (PNTZ) and operated in two different BN-600 fuel assemblies. Specimens of eight claddings irradiated at different temperatures to different damage doses have been examined. Maximum damage dose is 77 dpa.

Claddings from the second lot were manufactured at Public Joint Stock Company "Mashinostroitelny Zavod" (MSZ) according to the advanced technology. Fuel pins were operated in three different BN-600 fuel assemblies to maximum damage dose in the range between 84 and 96 (in different assemblies). Specimens of sixteen claddings irradiated at different temperatures to different damage doses have been examined.

Swelling values are determined on tubular specimens of claddings by hydrostatic weighing. Density of cladding material is determined with Sartorius balance equipped with YDK01 density determination kit. Ethanol is used as an auxiliary liquid. 30...31 mm long pieces of claddings with mechanically removed fuel are taken as specimens. Specimen preparation for weighing is made by chemical etching of fuel composition residuals. Density is calculated by the formula [2]

$$\rho = \frac{M_A \times (\rho_L - \sigma)}{0,99983 \times (M_A - M_L)} + \sigma, \quad (1)$$

where ρ is specimen material density, g/cm^3 ,

ρ_A is liquid density, g/cm^3 ,

M_A and M_L are balance readings for specimen weighing in air and liquid, respectively, g,

$\sigma = 0,001173 \text{ g/cm}^3$ is typical value for density of dry atmospheric air at pressure of 740 mm Hg and temperature of 20 °C.

0.99983 is an adjustment for measurement error for the force of bouyancy from a sample holder immersed into liquid (in YDK01 kit using $\varnothing 76$ mm beaker and $\varnothing 0.7$ mm wires).

Ethanol density is measured with the glass plummet from YDK01 kit. Ethanol temperature during examination is in the range between 23 and 26 °C.

Radiation-induced swelling S is calculated according to density determination results by the formula

$$S = \frac{\rho_0 - \rho}{\rho} \cdot 100 \%, \quad (2)$$

where ρ_0 is initial density of specimen material, g/cm^3 .

Accuracy of density determination by hydrostatic weighing is 0.2 %.

All the specimens are divided into groups correlating with irradiation temperature intervals (10 °C step size). This enables obtaining swelling-damage dose relation for different irradiation temperatures. Also specimens from some temperature intervals with close irradiation doses are examined with electron microscopy (TEM) to identify specific features of radiation void characteristics.

3. Results and Discussion

According to many studies, as damage dose of neutron irradiation increases, so does the swelling. This nonlinear process can be divided into three consecutive stages [3]:

1. incubation, when there is almost no swelling,
2. unsteady-state swelling, when swelling increases along with dose rate (swelling rate).
3. steady-state (stationary) swelling, when swelling rate is constant.

The first stage is characterized by incubation dose D_0 , marking the beginning of the second stage. The beginning of swelling is considered to be the moment when swelling is $S_0=0.1\%$ or $S_0=1.0\%$, which is experimentally determined relative value S_0 .

No quantitative characteristic has been introduced to mark the ending of the second stage.

The third stage is distinguished by the value of stationary swelling rate. Some works suppose this 'universal constant' to be $1\%/dpa$ for all austenitic steels [4-6]. However other studies show stationary swelling stage to be of other values in particular cases [7, 8].

The paper gives two result arrays: No. 1 – data for EK-164 claddings by PNTZ, and No. 2 – data for EK-164 claddings by MSZ, manufactured according to the advanced technology. Damage dose dependence of experimentally measured swelling for different irradiation temperatures (divided into intervals with $10\text{ }^\circ\text{C}$ step size) is defined. These relations are given for different temperature intervals (*see Fig. 1...6*). Maximum swelling values registered for EK-164 claddings by MSZ at damage doses of $\sim 94\text{ dpa}$ are $\sim 8...9\%$ (*see Fig. 4*). As it was shown in studies [9, 10], during examination of ChS-68 austenitic steel steady-state swelling begins at swelling of $\sim 9\%$. Therefore it can be concluded that swelling is within the unsteady-state stage for the whole studied temperature and dose range. It also should be noted that swelling values vary significantly within temperature intervals at close irradiation doses. It is clear that in a wide temperature range between $410\text{ }^\circ\text{C}$ and $570\text{ }^\circ\text{C}$ (*see Fig. 1...5*) swelling of claddings made of EK-164 steel by PNTZ is greater than that for claddings by MSZ. It confirms that improvement of EK-164 steel manufacturing technology ensured increase of its radiation resistance. At irradiation temperatures above $580\text{ }^\circ\text{C}$ swelling of EK-164 cladding specimens, determined by hydrostatic weighing, almost does not depend on irradiation dose and manufacturing technology and is up to 1.5% (*see Fig. 6*).

Specimens have been examined by electron microscopy (TEM) in a wide irradiation temperature range between 380 and $610\text{ }^\circ\text{C}$. As it was shown in studies [11, 12], at all irradiation temperatures and doses in material structure there are uniformly distributed small voids, average size of $2...3\text{ nm}$, located predominantly on dislocation structural elements along grain boundaries (*see Fig. 7*), and intergranular and grain boundary second phase precipitates (*see Fig. 8*). Types and number of second phase precipitates varies depending on irradiation temperature.

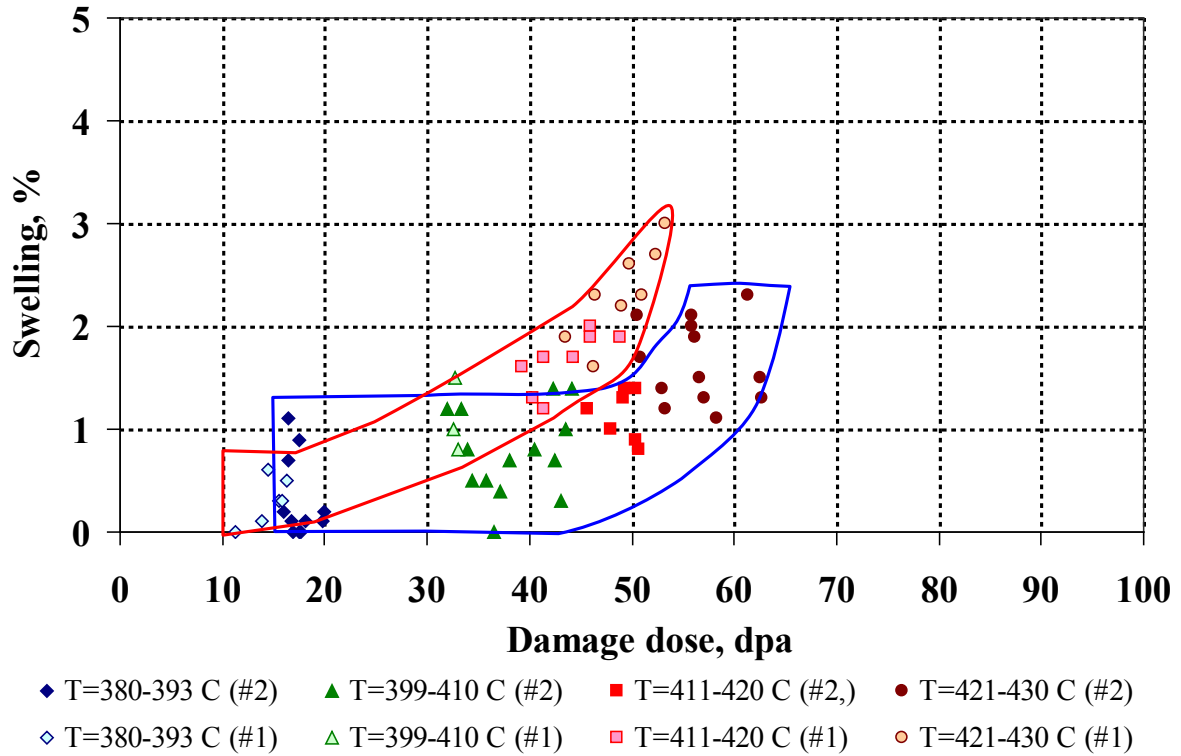


FIG. 1. Swelling dependence of cold-worked EK-164 (IAR) steel by PNTZ ((#1) and MSZ (#2) on damage dose for 380...430 °C irradiation temperature interval.

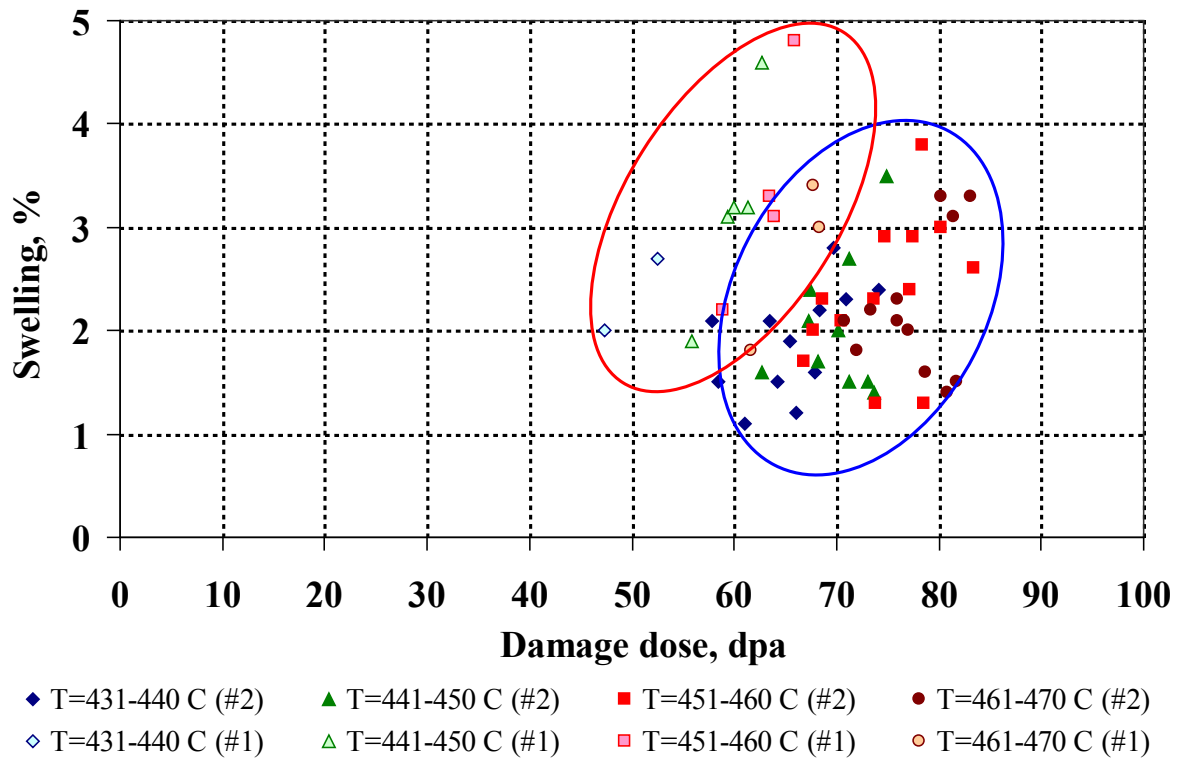


FIG. 2. Swelling dependence of cold-worked EK-164 (IAR) steel by PNTZ ((#1) and MSZ (#2) on damage dose for 431...470 °C irradiation temperature interval.

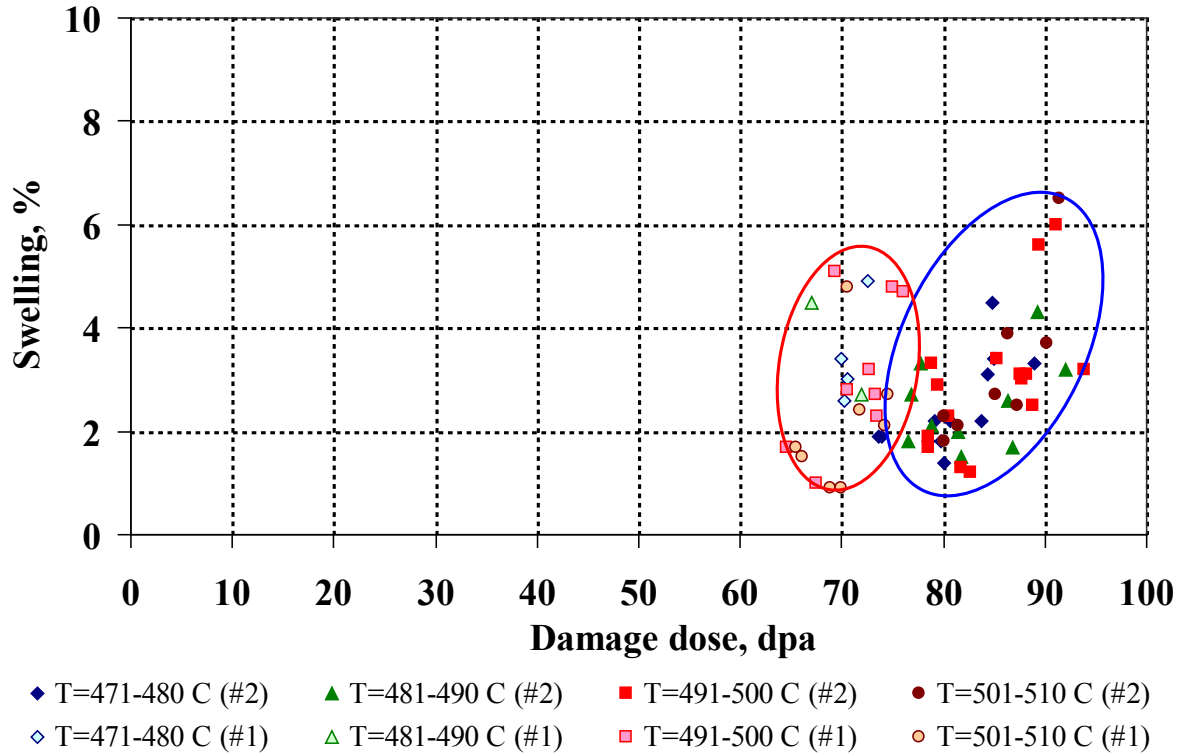


FIG. 3. Swelling dependence of cold-worked EK-164 (IAR) steel by PNTZ ((#1) and MSZ (#2) on damage dose for 471...510 °C irradiation temperature interval.

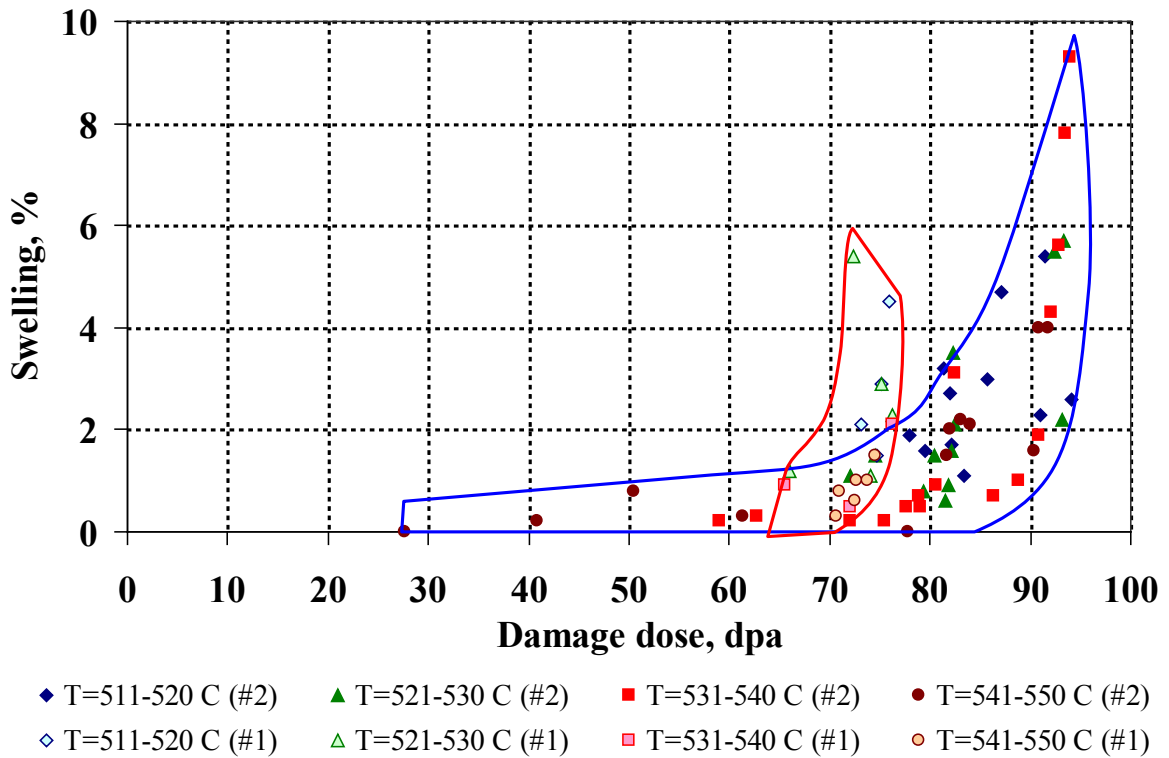


FIG. 4. Swelling dependence of cold-worked EK-164 (IAR) steel by PNTZ ((#1) and MSZ (#2) on damage dose for 511...550 °C irradiation temperature interval.

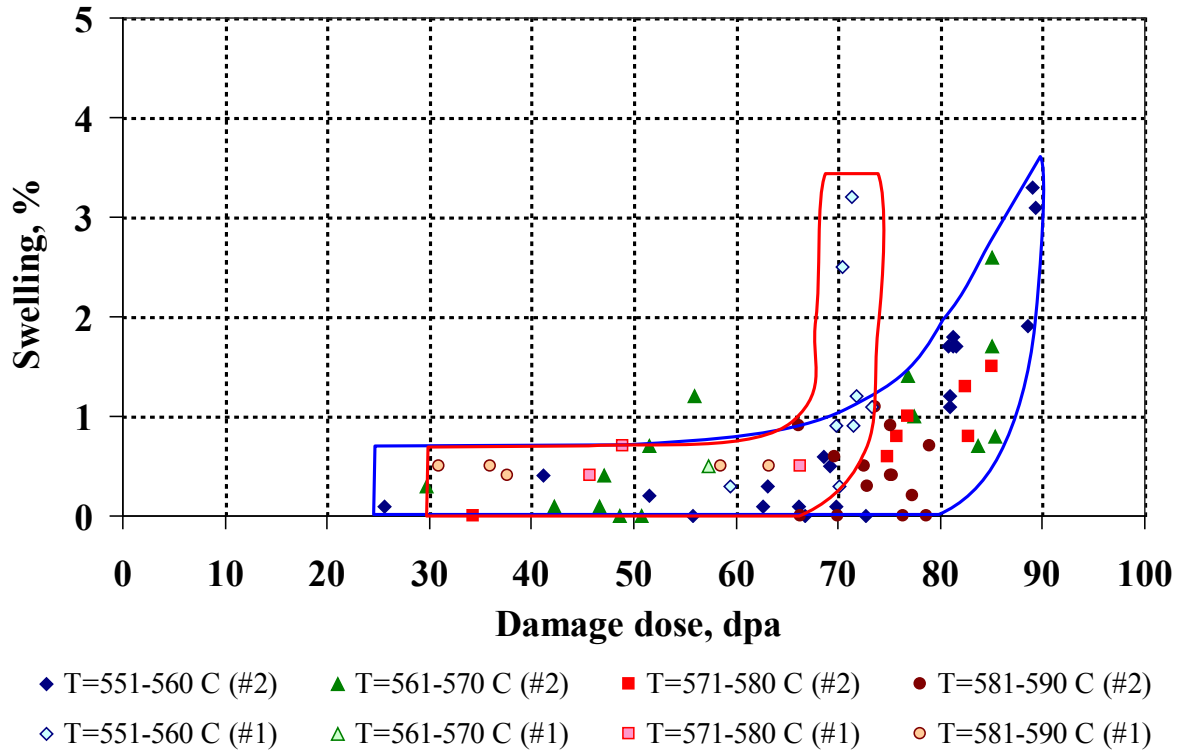


FIG. 5. Swelling dependence of cold-worked EK-164 (IAR) steel by PNTZ ((#1) and MSZ (#2) on damage dose for 551...590 °C irradiation temperature interval 551 and 590 °C.

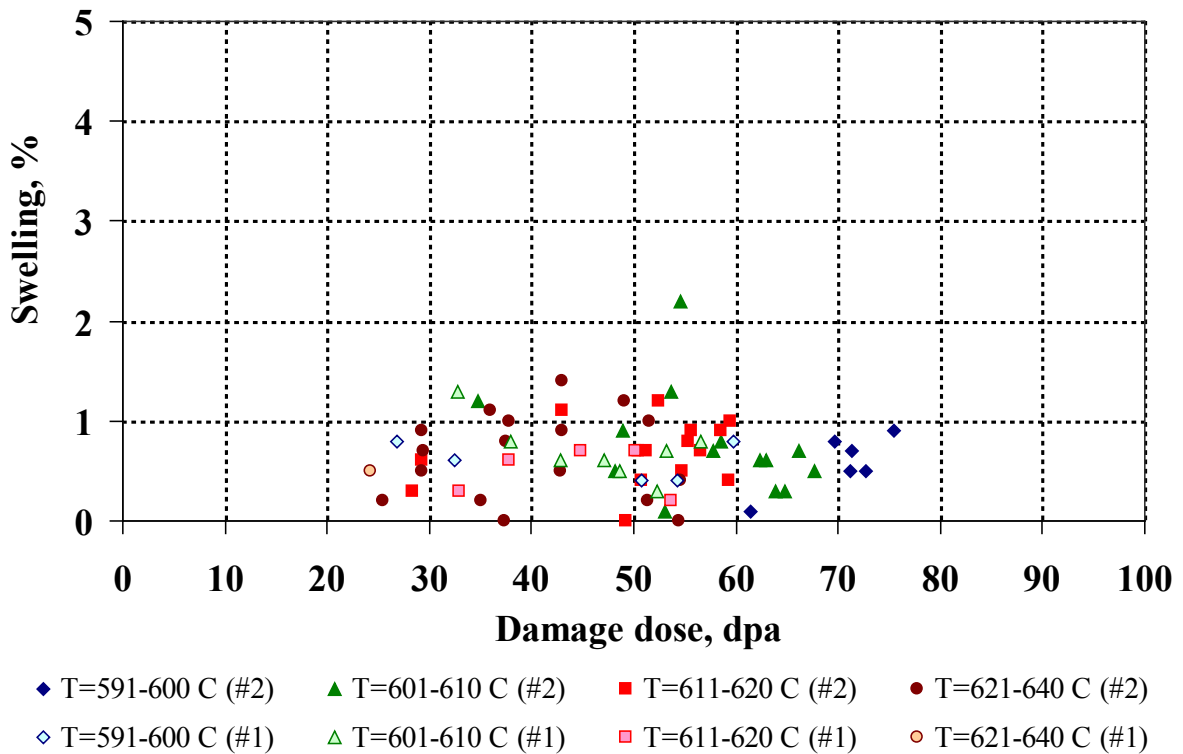


FIG. 6. Swelling dependence of cold-worked EK-164 (IAR) steel by PNTZ ((#1) and MSZ (#2) on damage dose for 591...640 °C irradiation temperature interval.

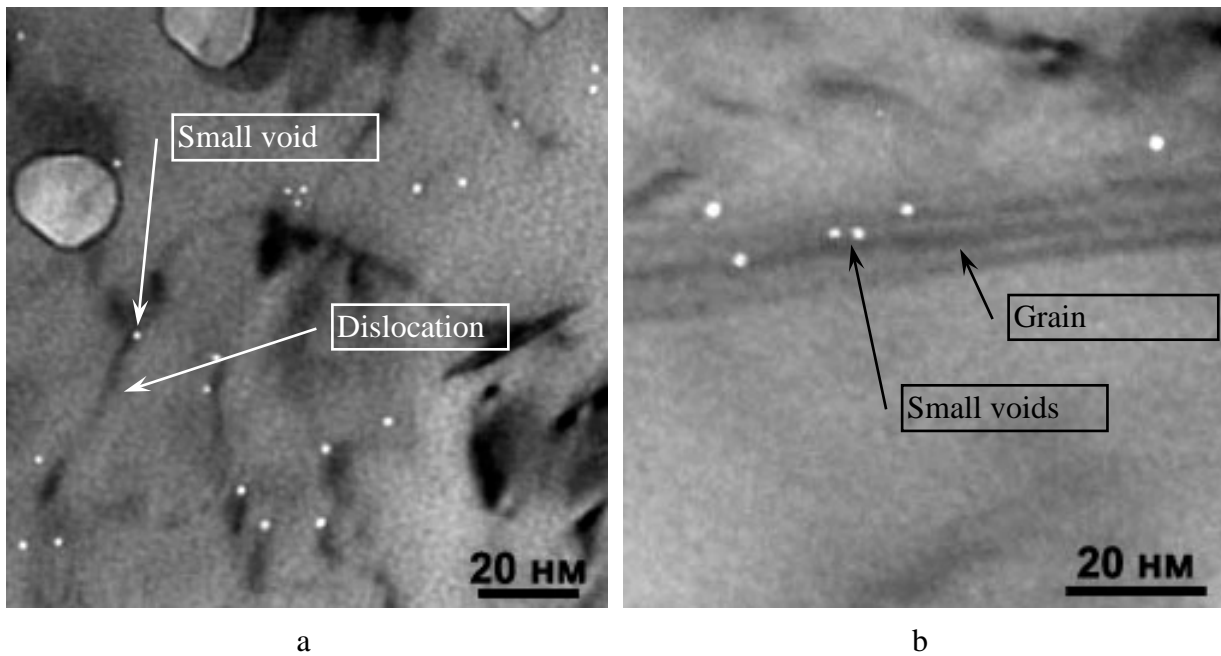


FIG. 7. Small voids on dislocations (a) and grain boundaries (b) in cold-worked EK-164 (IAR) steel after irradiation at 445 ± 5 °C [12].

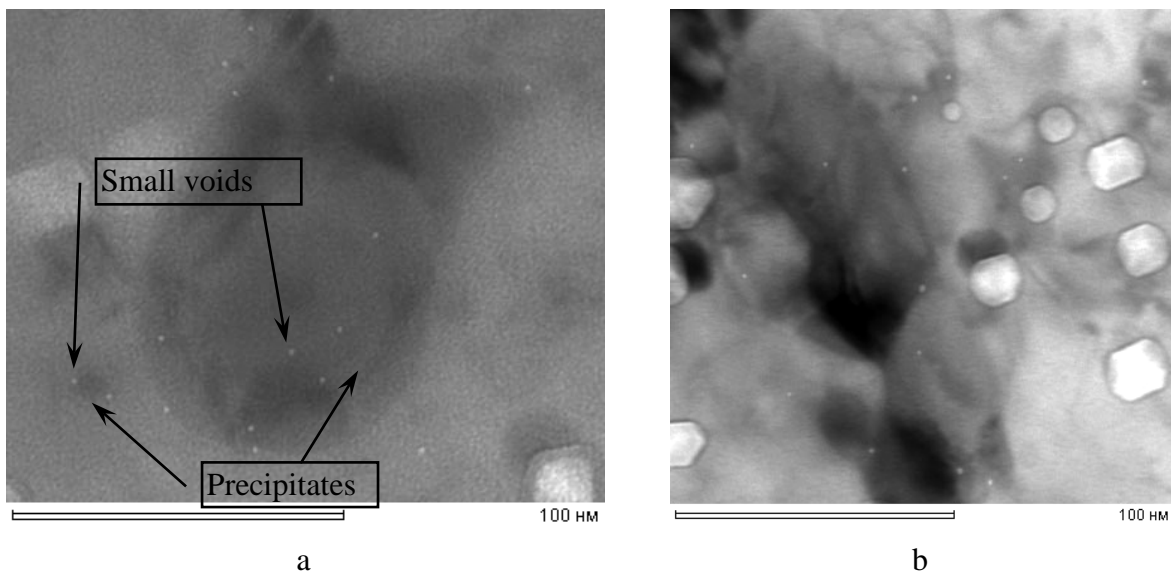


FIG. 8. Small voids on intergranular (a) and grain boundary (b) second phase precipitates in cold-worked EK-164 (IAR) steel after irradiation at 45 ± 5 °C.

At irradiation temperature of 385 ± 5 °C, apart from small voids, larger voids of 9...10 nm on average appear. As irradiation temperature and dose increase, size range for these voids shifts towards larger sizes (see Fig. 9), and their distribution by volume becomes less uniform (see Fig. 10). Small voids, despite of their high concentration, do not significantly contribute to material swelling.

To identify differences in porosity characteristics vacancy void parameters, obtained for EK-164 specimens by PNTZ and MSZ after operation in low enrichment zone of BN-600 reactor, are compared, as they have been operated in reactor for similar periods of time. Damage dose dependence of average sizes and large void concentrations (as they significantly contribute to material swelling) in specimens are shown, correlating to 10°C-step ranges: low-temperature

(440...450 °C), intermediate-temperature (480...490 °C and 510...520 °C), and high-temperature (590...600 °C) (see Fig. 11).

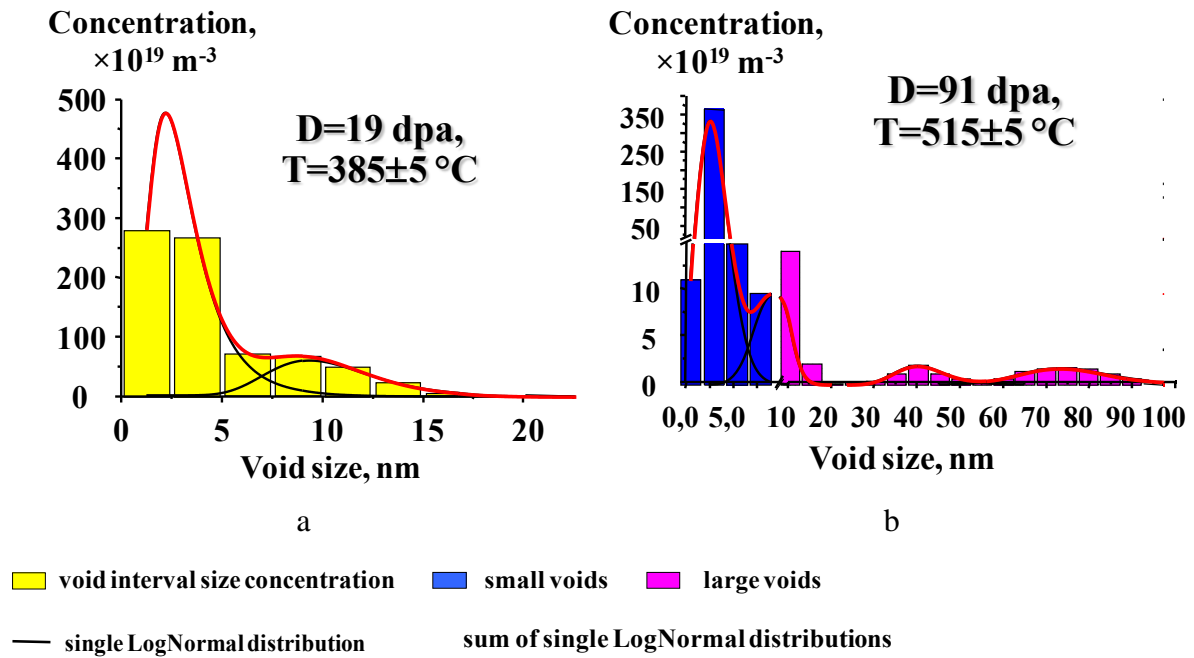


FIG. 9. Typical bar charts of void distribution by size in cold-worked EK-164 (IAR) steel after irradiation at $385 \pm 5 \text{ }^\circ\text{C}$ (a) and $515 \pm 5 \text{ }^\circ\text{C}$ (b).

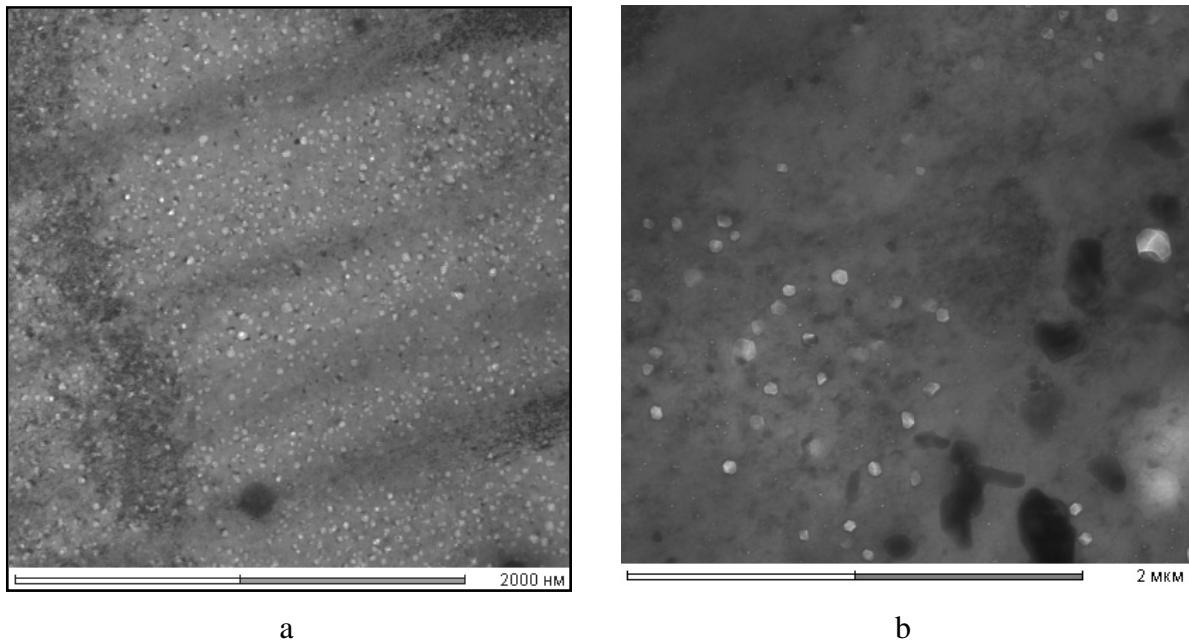
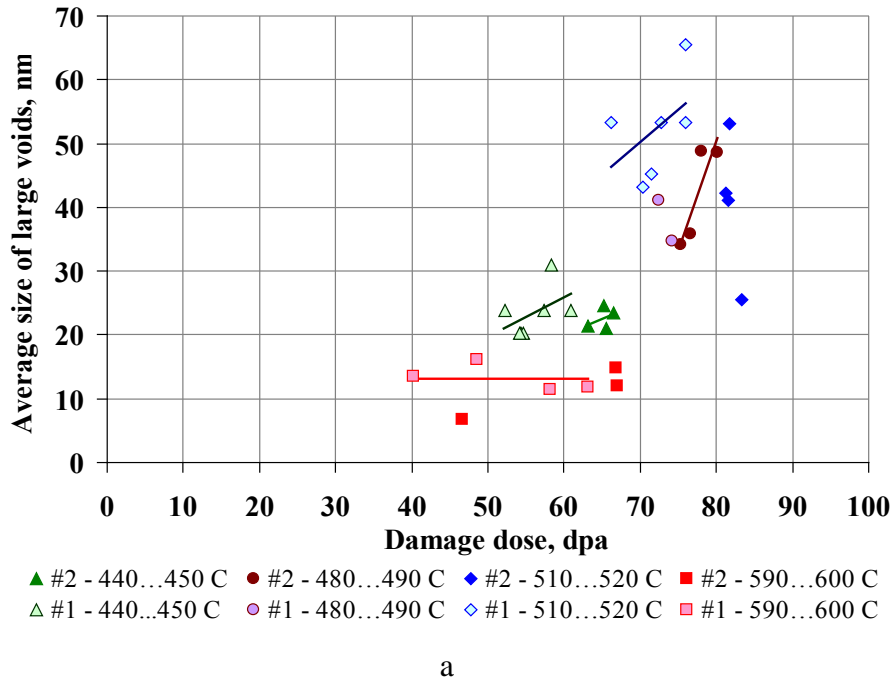
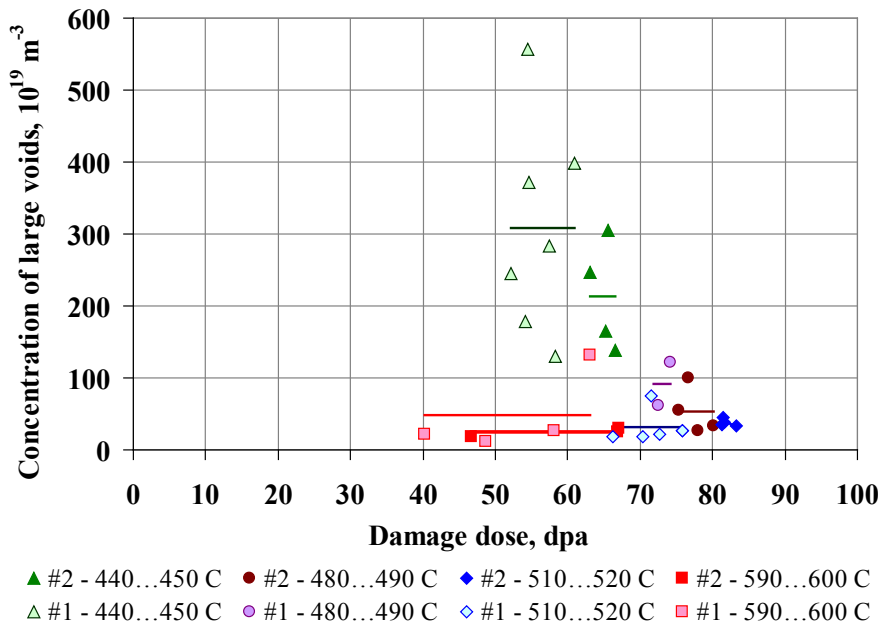


FIG. 10. Void distribution by volume in cold-worked EK-164 (IAR) steel material after irradiation at $445 \pm 5 \text{ }^\circ\text{C}$ (a) and $515 \pm 5 \text{ }^\circ\text{C}$ (b).

It appears that for low-temperature and intermediate-temperature intervals average size of large voids tends to increase along with damage dose, with absolute values for average size greater in material by PNTZ than in that by MSZ (see Fig. 11a).



a



b

FIG. 11. Dependence of large void average size (a) and concentration (b) in EK-164 steel by PNTZ (#1 – empty labels) and MSZ (#2 – filled labels) on damage dose for different irradiation temperature intervals.

In high-temperature interval average size for voids of both types does not vary and is at the same level for both steel materials (see Fig. 11a). There is no dependence of large void concentration on damage dose in any temperature interval, but large spreads of values are detected (see Fig. 11b). Therefore void concentrations can be averaged for each temperature interval (marked with straight lines in charts). It should be noted that in low-temperature and intermediate-temperature (480...490 °C) intervals averaged concentrations of large voids are greater for the material by PNTZ than by MSZ (see Fig. 11b). At higher irradiation

temperatures averaged values for large void concentrations are compatible irrespective of irradiation temperature and EK-164 steel manufacturing technology (*see Fig. 11b*).

Maximum void concentrations appear in temperature interval between 440 and 450 °C, but void size in this range does not exceed 50...60 nm. Thus swelling values vary slightly (*see Fig. 2*). With irradiation temperature increasing large void concentration reduces sharply. However, their size range increases to 100 nm, thereby increasing swelling values as well (*see Fig. 4*). At irradiation temperatures above 580 °C large voids in material structure decrease in number, small voids of high concentration appear, and estimated with TEM swelling does not exceed tenths of a percent. Therefore swelling values for high-temperature specimens by hydrostatic weighing are excessive. Probably this is related to significant changes in phase composition of specimens in this temperature interval, as well as to corrosion damages from the internal surface.

Nowadays MSZ in cooperation with VNIINM has completed the second phase of improvement of cladding manufacturing technology. Fuel pins with these claddings have been operated in BN-600 reactor to damage doses of ~100 dpa. Scheduled post irradiation examination of fuel pins will enable more precise estimation of fuel pins with EK-164 claddings.

4. Conclusion

The results of swelling determination for EK-164 claddings by hydrostatic weighing are given.

Swelling-dose dependence for different irradiation temperature intervals is shown.

It is found that with improved manufacturing technology for EK-164 claddings their resistance to radiation-induced swelling increases.

TEM investigation of radiation porosity establishes that from temperatures of 380...390 °C and doses of ~20 dpa small voids (2...3 nm in size) appear in steel.

With dose and temperature increase on bar chart of void distribution by size, maximum of larger voids, along with maximum of 2...3 nm voids, is detected.

Further temperature increase shifts large void maximum to larger sizes and reduces their concentration, and spatial distribution of voids becomes less uniform.

As damage dose increases, so does the average size of large voids, and absolute values in EK-164 steel specimens by PNTZ are greater than those for the material by MSZ up to irradiation temperatures of 510...520 °C. At temperatures of 590...600 °C they do not vary, irrespective of dose or manufacturing technology.

There are big spreads of large void concentration inside irradiation temperature intervals, irrespective of damage dose, and absolute values in EK-164 steel specimens by PNTZ are greater than those for the material by MSZ up to irradiation temperatures of 480...490 °C. At higher temperatures large void concentration is the same for materials of both manufacturing technologies.

According to the results of swelling investigation, examined fuel pins with EK-164 claddings can ensure service life to damage doses of at least 105 dpa.

5. References

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