Fracture strain and fracture toughness prediction for irradiated austenitic steels over wide range of temperatures taking into account the effect of swelling and thermal ageing

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Abstract. The work considers two mechanisms of embrittlement of austenitic steels under irradiation over wide range of temperatures result in quasi-brittle intergranular and ductile transgranular fracture. Model is developed for prediction of both quasi-brittle intergranular and ductile transgranular fracture and the fracture mechanisms transition. The model allows one to predict fracture strain and fracture toughness of material for different stress triaxiality taking into account the influence of neutron radiation, swelling and grain boundary damage by He. The data are represented of fracture modeling for the material of decommissioned fuel assemblies.

Key Words: Fast reactor, Embrittlement and damage mechanisms, Fracture models

1. Introduction

Fracture toughness and fracture strain of austenitic steels are the important performance properties, which control serviceability of the components of fast reactors including fuel assemblies. It is known that the above properties decrease under irradiation and thermal ageing. Especially strong decrease of fracture toughness occurs under irradiation accompanied by swelling.

It is necessary to note that ductile transgranular fracture mechanism dominates at temperature less than 500°C even for highly embrittled material with high swelling [1]. Such type of fracture can be predicted by the ductile fracture model proposed by authors early [2, 3]. At the temperature higher than 500°C fracture of irradiated austenitic steels occurs by intergranular mode. Such fracture is connected with embrittlement known as high temperature radiation embrittlement (HTRE) and caused by a decrease in the strength of grain boundaries due to growth of helium (He) pressure with an increase in temperature. There are only few experimental data on HTRE [4], and models are practically absent for prediction of fracture toughness and ductility for different levels of stress triaxiality.

Some components of BN-type fast neutron reactors are undergo high dose of neutron irradiation (D > 45 dpa) at elevated operation temperatures (T>450-500°C). In the first place, these components are fuel cell shells and neutron reflector made from austenitic steel.

Combination of elevated irradiation temperature with high neutron dose results in embrittlement of austenitic steels by both above mechanisms. Depending on specific combination of irradiation dose and temperature, the component strength can be controlled by one of the above mechanisms.

The aim of the paper is to develop and to present models allowing one to predict ductility and fracture toughness of irradiated austenitic steels with regard for different embrittlement mechanisms. Such prediction allows one to estimate strength of BN-type fast reactor component for which material degradation is most strong.

2. Transgranular ductile fracture

The features of ductile transgranular fracture of irradiated austenitic steels are considered in detail in papers [2, 3, 5]. In these papers the ductile fracture model is presented which allowing one to predict ductility and fracture toughness of irradiated austenitic steels with regard for the effect of swelling.

The main considerations of the developed model are the following.

Fracture proceeds by the mechanism of nucleation, growth and coalescence of voids. Two void populations are considered: vacancy voids and deformation voids. Vacancy voids are nucleated under irradiation resulting in swelling and deformation ones are nucleated on inclusions and impurities [2] under plastic deformation.

The deformation void concentration rate $\frac{d\rho_v^{def}}{d\sigma_{nuc}}$ is presented in the form [2]

$$\frac{d\rho_v^{def}}{d\sigma_{nuc}} = \frac{\rho_v^{max} - \rho_v^{def}}{\sigma_d}, \tag{1}$$

where ρ_v^{def} is the concentration of deformation voids per unit volume of the material, ρ_v^{max} is the maximum concentration of void nucleation sites, or other words maximum concentration of inclusions, σ_{nuc} is the stress resulting in void nucleation; σ_d is the effective local strength of matrix-inclusion interphase. In the general case the parameter σ_d depends on the particle shape.

In Eq. (1) σ_{nuc} is the stress controlling nucleation of discontinuity near some barriers and, in particular, nucleation of voids or microcracks near some inclusions [6, 7]. An equation describing the stress of cleavage microcrack initiation [6, 7] has been derived from the analysis of stresses near the head of dislocation pile-up

$$\sigma_{nuc} = \sigma_1 + m_{T\varepsilon} \cdot \sigma_{eff} \tag{2}$$

where the parameter $m_{T\varepsilon}$ is the coefficient of local stress concentration near dislocation pile-ups. Generally, $m_{T\varepsilon}$ depends on test temperature and plastic strain; σ_1 is the maximum principal stress; $\sigma_{eff}=\sigma_{eq} - \sigma_Y$ is the effective stress; σ_{eq} is the equivalent stress; σ_Y is the yield stress.

When analysing the growth of vacancy and deformation voids the Huang equation [8] is used. This equation has been modified for the case when the distance between voids is comparable with their sizes. It is assumed the rate of void growth increases due to an additional strain concentration in the vicinity of voids. To describe the voids growth allowing for their interaction an additional factor in the form of $\frac{1}{1-f}$ was introduced into the Huang equation

$$\frac{dV_{void}}{V_{void}} = \frac{3 \cdot \alpha}{1-f} \cdot d\bar{\varepsilon}_{eq}^{p}. \tag{3}$$

where f is the void volume fraction in a material; α is the coefficient depending on stress triaxiality (ST) $\frac{\sigma_m}{\sigma_{eq}}$, α increases with increase of ST; $\bar{\varepsilon}_{eq}^{p} = \int d\varepsilon_{eq}^{p}$ is the Odquist parameter; $d\varepsilon_{eq}^{p}$ is the equivalent of a plastic strain increment

The criterion of a unit cell plastic collapse or, in other words, the criterion of plastic instability is used as fracture criterion and formulated as [9]

$$\frac{dF_{eq}}{d\bar{\varepsilon}^{p}_{eq}} = 0\,, \tag{4}$$

where $F_{eq} = \sigma_{eq}\cdot(1 - \overline{A}_{\Sigma})$; $\overline{A}_{\Sigma}$ is the relative area of voids, i.e. the total area of voids on the unit area of unit cell with voids. It should be noted that when analysing Eq. (4) the stress triaxiality is taken to be constant [9].

From Eqs. (2) and (3) it is seen that an increase of ST results in additional increase of σ_{nuc} and α. Hereupon with increase of ST more intensive voids nucleation and growth happens that results in a decrease of fracture strain for ductile fracture $\varepsilon_{f_{ductile}}$

Let's consider the effect of neutron irradiation on ductile fracture and a decrease of ductility from point of view of the presented model.

In the first place, the neutron irradiation leads to the strong increase of yield strength and ultimate strength and also results in decrease in strain hardening of a material. These processes are connected with formation of large amount of point defects and dislocation loops under irradiation.

Radiation hardening of the material leads to an increase in principle stress σ_1 under loading and therefore to the increase in nucleation stress σ_{nuc}. Therefore, more number of voids is nucleated in irradiated material than in unirradiated one with the same amount of inclusions. It is necessary to note that neutron irradiation without thermal ageing does not affect on maximum concentration of void nucleation sites ρ_v^{max}. At elevated temperature more than 450°C the carbides are generated which can be sites for deformation voids nucleation. Thus in general case at thermal ageing accompanied by carbide formation the value of ρ_v^{max} is increased.

The increase of deformation voids concentration is connected also with decrease in interphase strength σ_d. A decrease in σ_d is connected with several mechanisms, in particular, with the segregation of different impurities at the interphase boundaries and with the internal stress at these boundaries due to dislocation loops formation [6, 7, 10]. These processes result in the weakening inclusion-matrix interphase. In addition, σ_d decreases due to an easier formation of dislocation pile-ups near void nucleation sites under irradiation.

Third cause of the decrease in ductility under irradiation is a decrease in the strain hardening of a material mentioned above. According to criterion (4) the decrease in strain hardening results in voids coalescence at lower plastic strain when other parameters are fixed. \

In frame of verification of ductile fracture model it is shown in paper [2] that dependence $\varepsilon_{f_{ductile}}$ (T_{test}) for irradiated material is basically controlled by strain hardening and its dependence on temperature.

In addition, the ductile fracture model described above allows one to predict effect of neutron irradiation on fracture toughness taking into account the effect of stress triaxiality on fracture strain near the crack tip. Fracture toughness decreases under neutron irradiation much stronger in comparison with the fracture strain of smooth tensile specimens due to higher stress triaxiality near the crack tip. According to paper [11] for 18Cr-10Ni-Ti grade steel (Russian analog of AISI 321 steel) maximal decrease of the fracture strain under irradiation without swelling and thermal ageing reaches 2 times. In the same time fracture toughness decreases by 5-7 times [11, 12].

We should also consider the effect of radiation swelling on ductility and fracture toughness under ductile fracture. Processes of growth and coalescence of vacancy voids under deformation are similar to ones for deformation voids and hence these processes may be described by the same equations. Experimental data and calculations with the presented ductile fracture model show a decrease of the fracture strain for smooth tensile specimens up to 3 times at ~6% swelling in comparison with irradiated material without swelling [2].

Unlike the fracture strain, the fracture toughness decreases much stronger with increase of swelling. It is connected with singularity of stress-strain fields near the crack tip and the existence of so-called process zone, i.e. the area in which elementary act of fracture occurs [2, 3, 5].

The process zone size r_f for classical ductile fracture of material without swelling as a first approximation depends on distance between the void-nucleating inclusions [13, 14]. In this case, process zone size is about ~100 μm. With swelling growth, vacancy voids spaced tens of nanometers begin to dominate in ductile fracture. It results in the decrease of r_f by 3 order when swelling increases to 5-7%. In paper [2] it is shown by experimental and calculative method that value of r_f decreases from 100 μm for material without swelling to ~0.3 μm for material with ~5% swelling. This process leads to sharp decrease of J_c down to values close to zero (See Fig. 1).

3. Intergranular fracture by HTRE mechanism

When test temperature for irradiated austenitic material is above 500° the fracture mechanism in many cases changes from the transgranular ductile to intergranular quasibrittle fracture. The fracture mechanism change is connected with so-called high-temperature radiation embrittlement (HTRE) caused by a decrease in the strength of grain boundaries due to higher helium pressure with an increase in temperature. Helium is generated from nuclear reactions through the interaction of neutrons with such elements as Ni, B and Fe. The mechanism HTRE leads to a sharp decrease in fracture strain and fracture toughness at temperatures above ≈500°C.

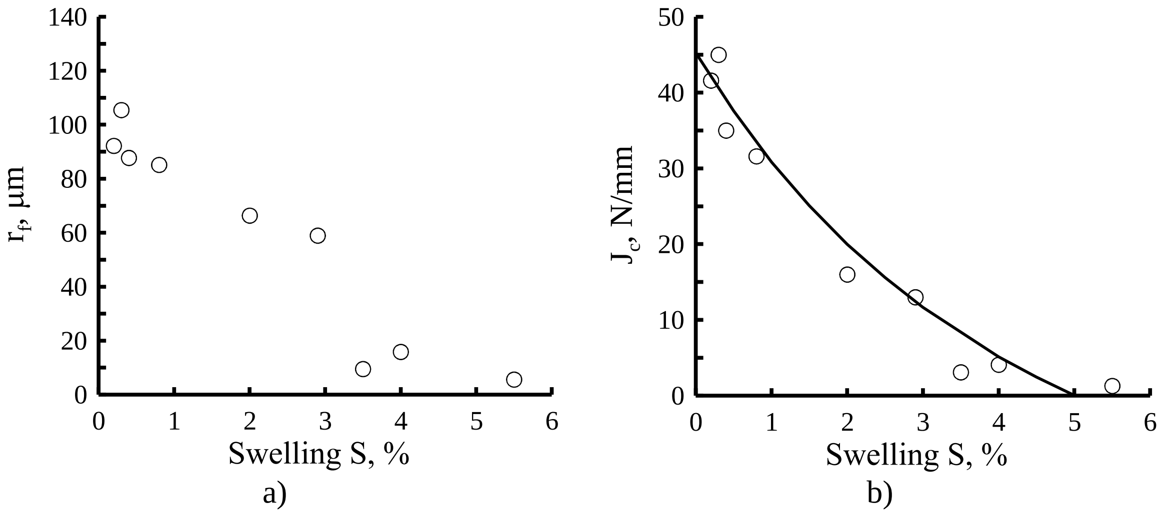


FIG. 1. Effect of the swelling S on the process zone size r_f and fracture toughness J_c for the material of BOR-60 reactor shield assembly irradiated over the dose range from 90 to 150 dpa and tested at 290°C; curve is a prediction by ductile fracture model

In spite of the fact that this fracture mechanism was revealed as far back as 1960s, there is no fracture criterion for this mechanism as applied to the structural integrity of reactor components till now. In order to formulate fracture criteria describing this mechanism the special experiments were carried out as well as the results of testing BN-600 reactor fuel cell shells were used. For the effect of stress triaxiality on fracture strain to be determined specimens of 2 types were tested: smooth cylindrical specimens and notched cylindrical specimens. For smooth specimens the maximum triaxiality $\sigma_m/\sigma_{eq} \approx 0.5$, for notched specimens the maximum triaxiality $\sigma_m/\sigma_{eq} \approx 1$.

Figure 2 shows the temperature dependences of fracture strain, $\varepsilon_f(T_{test})$, for specimens of both types (the presented results have been obtained together with Ph.D. O. Prokoshev). As is seen from the figure, at a temperature of 550-600°C when fracture proceeds by the HTRE mechanism only, the fracture strain for both types of specimens is nearly the same. Therefore, the fracture strain for the HTRE mechanism unlike ductile transgranular fracture is invariant with respect to stress triaxiality.

The result obtained is not obvious since generally stress triaxiality affects brittle and quasi-brittle fracture. This result seems to be explained as follows. Grain boundaries are usually permeable barriers for moving dislocations unlike carbides or other strong particles where the initiation of cleavage microcracks takes place [6].

At the same time, due to a different grain-boundary orientation the propagation of dislocations through grain boundaries results in formation of grain-boundary dislocations [15, 16]. With increasing plastic deformation, that is with increasing the number of dislocations moving through the grain boundary, the number of grain-boundary dislocations increases that finally generates considerable local stresses resulting in the formation of grain-boundary microcracks. When the test temperature grows an increasing disjoining pressure of He on grain boundaries makes easier the formation of grain-boundary microcracks. Second factor making easy the nucleation of grain-boundary microcracks is grain-boundary carbides formation (usually $Cr_{23}C_6$) due to thermal ageing. These carbides decrease grain boundary cohesive strength.

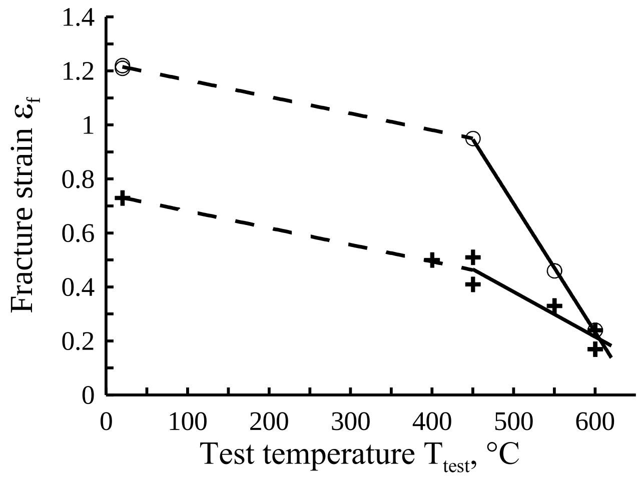


FIG. 2. Dependence of fracture strain ε_f on test temperature T_{test} for specimens with different triaxiality from 18Cr-10Ni-Ti grade steel irradiated up to ~1 dpa at 500°C during 123456 h: open dots correspond to smooth cylindrical specimens (σ_m/σ_{eq}≈0.5), crosses correspond to notched cylindrical specimens (σ_m/σ_{eq}≈1); dotted lines corresponds to ductile fracture; solid lines corresponds to transition from ductile fracture to intergranular fracture.

Local stresses from grain-boundary dislocations on the grain boundary are by far higher than the applied (nominal) ones. Therefore, stress triaxiality has a very slight effect on the initiation of grain-boundary microcracks, and the plastic strain $\varepsilon_{f_{He}}^{nuc}$ can be used as a criterion for grain-boundary microcrack nucleation.

The further grain-boundary microcrack propagation is like the brittle microcrack propagation [6, 7]. This propagation does not require high energy since grain boundaries are already weakened by helium, and the field of local high stresses follows the moving crack tip.

Therefore, HTRE differs from ductile fracture. The energy for crack nucleation under ductile fracture is close to energy of crack propagation. That's why for ductile mechanism crack propagation is a successive ductile fracture of small volume of material (unit cell) near the moving crack tip.

Thus, when the HTRE mechanism acts the condition of grain-boundary crack propagation can be written as the Griffith condition [17]

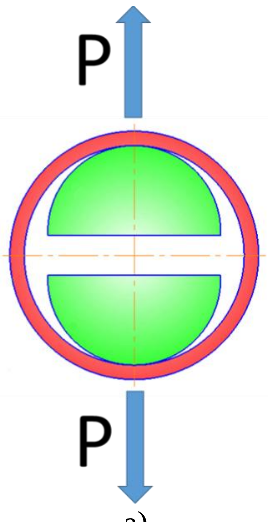
$$G = 2\gamma_{gb}, \tag{5}$$

where γ_{gb} is the effective surface energy of a grain-boundary cracks decreasing with an increase in temperature due to the disjoining helium effect.

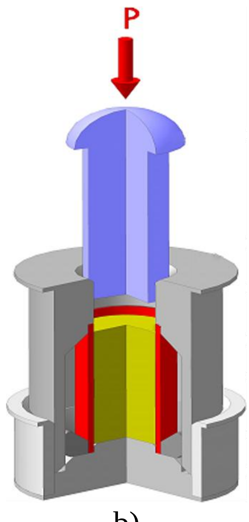
Let us consider if the above formulated criteria describe the observed fracture modes in testing specimens of two different types machined from BN-600 reactor fuel cell shells. The figure 3 shows two schemes of loading for the two type of specimens: the ring loading by its tension with two grips (specimen of type 1) and the cylinder loading by the incompressible filler pressure (specimen of type 2).

The SEM examination of the fracture surface of specimens has shown as follows. For Scheme 1 the specimen fracture occurred entirely by the intergranular mechanism. For Scheme 2 the fracture initiation also proceeded by the intergranular mechanism (the intergranular surface size is 1-2 grains), then the fracture followed by the transgranular mechanism.

For the usability of the formulated criteria for the above specimens to be analyzed FEM calculations were made. It was assumed that for both the types of specimens the initiation of intergranular fracture occurs with the same strain. After the strain $\varepsilon_{f_{He}}^{nuc}$=2% had been achieved on the inside surface of specimens, a 10 μm edge crack was postulated for the specimen of each type and then the dependence G(Δl)=J(Δl) was calculated when assuming that the crack propagates with no plastic strain and with the fixed displacement of the grips (Scheme 1) and the filler (Scheme 2).



a)



b)

FIG. 3. Two schemes of loading the specimens machined from BN-600 reactor fuel cell shells: a) the ring loading by its tension with two grips (specimen of type 1); b) the cylinder loading by the incompressible filler pressure (specimen of type 2).

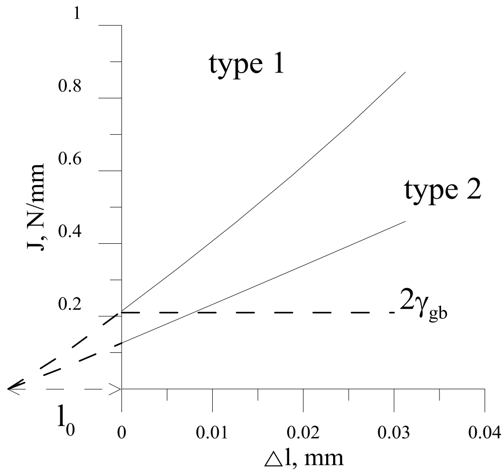


FIG. 4. Dependence of J-integral on crack propagation Δl for two types of specimens.

To exclude the plastic strain near the crack tip due to its propagation, the material yield strength just after the crack initiation was taken to be very large – equal to 10000 MPa.

The calculated dependences J(Δl) are represented in Fig. 4. As is seen from the figure, the dependence J(Δl) for the specimen of type 1 is steeper than that for the specimen of type 2. It is related to a higher energy capacity for the specimen of type 1.

If $2\gamma_{gb}$ has the level shown in the figure, the intergranular crack propagation in the specimen of type 1 after initiation will proceed without interruption to the specimen failure. In the specimen of type 2 after the crack initiation by the HTRE mechanism, the propagation will not be unstable since J(Δl) <$2\gamma_{gb}$. Therefore, further fracture of the specimen of type 2 can occur following two possible alternatives:

1. By the mechanism of the successive initiation of a grain-boundary microcrack with length of l_0 (See Fig.5a). For this variant additional loading of the specimen is required so that the strain $\varepsilon_{f_{He}}^{nuc}$ can be achieved over the zone Δl_1 equal to l_0.

2. By the ductile fracture mechanism (See Fig.5b). For this variant additional loading of the specimen is also required so that the strain $\varepsilon_{f_{ductile}}$ can be achieved over the zone Δl_1. Taking into account that the value $\varepsilon_{f_{ductile}}$ unlike $\varepsilon_{f_{He}}^{nuc}$ is sensitive to stress triaxiality (which is locally induced by the formation of a grain-boundary crack), the situation is quite possible when $\varepsilon_{f_{ductile}}$ is less than $\varepsilon_{f_{He}}^{nuc}$. For this case, the further crack propagation occurs by the ductile transgranular mechanism.

Thus, the proposed fracture criteria by the HTRE mechanism allow one to describe quite adequately the difference in fracture modes when testing specimens of different types made of the same material.

It should be noted that fracture modes for specimens of different types, apparently, can be nearly the same when specimens made of material with considerable swelling, for example, with the swelling S>5 %. The point is that at S>5 % the fracture toughness J_c for ductile fracture is very small due to the reduction of the so-called process zone till the sizes comparable to the distance between vacancy voids [2, 5]. If it proves to be that $J_c < 2\gamma_{gb}$, then after the initiation of an intergranular crack in the specimen of type 1 the intergranular fracture mode will change into the transgranular one. In the specimen of type 2 the change of the fracture mode will be similar to the specimen of type 1 in view of the following fact. If the condition $\varepsilon_{f_{ductile}} < \varepsilon_{f_{He}}^{nuc}$ is met at S=0, then this condition is all the more valid at the swelling S>0 since $\varepsilon_{f_{ductile}}$ decreases with increased swelling, and $\varepsilon_{f_{He}}^{nuc}$ does not depend on swelling because vacancy voids does no weak grain boundaries.

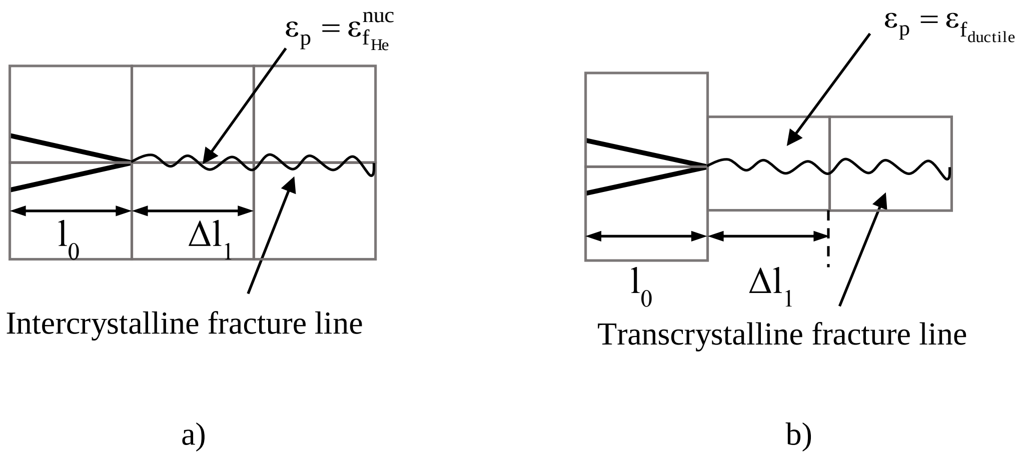


FIG. 5. Two variants of crack propagation in specimens of type 2: a) by the mechanism of the successive initiation of a grain-boundary microcrack; b) by the ductile fracture mechanism.

Thus, if we know parameters of ductile fracture model and parameters $\varepsilon_{f_{He}}^{nuc}$ and γ_{gb} of fracture model for HTRE it is possible to predict strength of BN-type reactor components under different neutron irradiation regimes with regard for stress-and-strain fields features.

4. Conclusions:

1) High doses of neutron irradiation at elevated temperatures (T ≥ 450°C) may result in considerable embrittlement of austenitic steels. Such embrittlement results in fracture by two main mechanisms: transgranular ductile fracture and intergranular quasibrittle fracture. The models have been proposed for prediction of ductility and fracture toughness of austenitic steels embrittled by mechanisms mentioned above.

2) Main considerations of ductile fracture model have been presented.

3) It is shown that fracture toughness may decrease in dozen times due to neutron irradiation and swelling even for ductile fracture

4) Main considerations of the proposed model of intergranular fracture for HTRE have been justified. It is shown that unlike the ductile fracture the condition of intergranular crack nucleation is invariant to stress triaxiality. The condition of crack propagation for HTRE also differs from one for ductile fracture. Crack propagation under ductile fracture is the successive initiation of ductile fracture near the tip of growing crack. Crack propagation under HTRE like Griffiths crack growth is described by energy criterion.

5) Combination of HTRE model with ductile fracture model developed early allows one to describe the parameters of fracture of irradiated austenitic steels over wide temperature range taking into account domination of different fracture mechanisms for different types of specimens and fracture mechanisms change with crack propagation in specimen of type 2.

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