РАЗДЕЛ ПЕРВЫЙ ФИЗИКА РАДИАЦИОННЫХ ПОВРЕЖДЕНИЙ И ЯВЛЕНИЙ В ТВЕРДЫХ ТЕЛАХ

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INFLUENCE OF ENVIRONMENTAL CONDITIONS ON MICROSTRUCTURE DEVELOPMENT AND SWELLING IN IRRADIATED AUSTENITIC STAINLESS STEELS

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Investigation and analysis of environmental conditions and swelling behaviour in irradiated austenitic stainless steels were done. Influence of dose rate, effects of stress and temperature history on microstructural evolution and swelling behaviour was studied in these steels.

INTRODUCTION

Possibility of achievement of high burn-up of fuel in Fast Breeder Reactor (FBR), which at this time define economy of reactors exploitation, and reactor's safety, now connected only with increasing of radiation stability of structural components (cladding and wrappers) materials. The most important problem to be solved in the increasing of radiation stability when the austenitic stainless steels are applied to the core materials of fast breeder reactors [1] is the suppression of void swelling under neutron irradiation.

As known [2] level of swelling is determined by balance of surviving point defects created during irradiation and which avoid mutual recombination and annihilation on any kinds of point defects sinks which or exist at material before irradiation or were created by irradiation. Any element of defect structure and any element of chemical composition in irradiated steels influence on sinks behaviour and diffusion properties of point defects under irradiation which as result generally determine swelling level.

Swelling problem in all types of reactors has additional difficulties: core structural materials work in complicated and, as a rule ,in non-stationary conditions (temperature, neutron flux, mechanical stress etc), which can exert a great distinctions in swelling behaviour.

It is mean application of materials in different irradiation environments requires not only understanding of the effects of irradiation on the microstructure, but also understanding how special specific environment conditions affects that microstructural development.

Spatial gradients in stress and displacement rate have all been found to be very important in determining the evolution of microstructure and macroscopic properties, including swelling behaviour and level. Impacts of temperature history during the early power-up stages clearly demonstrate the impact of such history on microstructural evolution [1]. Unfortunately up to now influence of changing these irradiation conditions on swelling behaviour in austenitic stainless steels are not sufficiently clear.

The successful strategies in material development for FBR must take into account difficulties arising from the radiation environment.

The purpose of this paper is to investigate and analyze influence of these factors on swelling level in irradiated austenitic stainless steels and select microscopic evolution responsible for such swelling behaviour.

ENVIRONMENTAL CONDITIONS AND SWELLING BEHAVIOUR IN IRRADIATED AUSTENITIC STAINLESS STEELS

Some structural components in FBR will work under neutron exposure greater than 100 dpa (displacements per atom). Under such bombardment and operating at liquid metal temperatures (300...700°C), most structural materials in the core of FBR are placed in a non equilibrium regime of microstructural alteration that is unprecedented in its ability to modify defect structure phase equilibria, change component dimensions.

Microstructural evolution in this temperature regime is very complex because it involves evolution of practically all components of microstructure:

- variable Frank loop formation and growth;
- variable RIS (radiation-induced segregation);
- variable formation and stability of radiation-enhanced/retarded -modified, or -induced precipitates;
- variable void and helium bubble formation and growth;
- variable dose (and dose rate) dependence of all above;
- strong temperature dependence of all above (with significant differences between 300...400°C and 400...600°C);
- strong coupling between many of the above at 400...
 600°C;
- strong sensitivity to alloy composition and pre-treatment of all above.

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Spatial changing in temperature (including temperature history), stress and displacement rate have all been found to be very important in determining these features of microstructure evolution and macroscopic properties.

DOSE RATE INFLUENCE

Dose rate or displacement rate (dpa rate) here will be described in units – dpa/sec.

Typically dpa rate in neutron experiments has a very wide region and can changes from 10^{-8} dpa/sec (in thermal reactors) to 5×10^{-6} dpa/sec (in active zones of fast breeder reactors).

The most of received results clearly demonstrate one tendency that swelling increases with decreasing dose rate (fig. 1) [1].



Fig. 1. Effect of displacement rate on the swelling-induced diameter change of annealed AISI 304 tubes at 390 ± 10 °C [1]

From physical point of view dpa rate providing a definition number of point defects and super saturation which is needed for voids formation and growth, due to changing of dpa rate irradiation can pay influence on processes of nucleation and growth of all components in steels microstructure: dislocation loops, voids, precipitates. Beside it dpa rate can modify rate of segregation processes and regulate decay of solid solution and precipitation in irradiated steels.

Early results of simulation experiments on charge particles accelerators results that the swelling rate was to be strongly dependent on temperature, "such temperature shifts" would produce large increases or decreases in swelling, depending on the temperature regime in which the irradiation was being conducted [3].

When it became apparent that the steady state swelling rate was not a strong function of irradiation temperature, it was realized that differences in displacement rate at most temperatures could only exert their influence on the duration of the transient regime of swelling. It was shown that dose dependence of void swelling in austenitic stainless steels is known to exhibit three regimes of response at a given displacement rate and temperature: the incubation period, the transient regime and the steady-state regime. Each of these regime has respond to differences in displacement rate [4]. For many austenitic steels, particular, 304 and 316 stainless steels, the incubation and transient regime are seems to decrease as the displacement rate is reduced, leading to earlier and therefore larger swelling. Fig. 2 demonstrates that relatively small differences in displacement rate have indeed been found to induce large changes in the duration of the transient regime at relatively high temperatures, but not in the post transient behaviour [5].

Results of work [6] showed a relative temperature shift in maximum swelling temperature at different dpa rate (fig. 3). Performed comparison of different damage rates shows that a temperature of maximum swelling is lower under a lower damage rate than at higher one. At the same time authors suggested that incubation swelling dose depends on neutron irradiation temperature and radiation damage rate and the incubation dose decreases with the reduction of these characteristics within tested temperature and dose rate interval.



Fig. 2. Influence of displacement rate on swelling of (left) SA AISI 316 fuel pin cladding at 562°C and (right) CW AISI 316 at 590...610°C [5]

First impression that defects super saturation and thereby void nucleation are predicted to be enhanced at higher displacement rates and therefore should produce shorter incubation periods.

At the same time experimental results, as was shown above, demonstrate that increases in displacement rate frequently produce longer incubation dose. It can be a consequence of several rate dependent processes, which are connected with high super saturation of point defects and increasing their diffusion mobility.

It is necessary to detach more sensitive to damage rate changing features of microstructural evolution: dislocations components, voids, precipitates or solid solution as result of different segregation possibilities at different dpa rate.





Fig. 3. Temperature dependence of swelling for samples of 16Crl5Ni2MolMn steel irradiated with different damage rate in BN-600 [6]

Dislocation structure serves as dispatcher in all further evolution of microstructure, because ever at small damage doses it became a dominant sink for point defects.

In approximation of homogeneous nucleation concentration of interstitials loops is expressed as follows:

$$N_{L} = N_{0} K^{1/2} \exp\left(E_{m}^{i}/2 kT\right), \qquad (1)$$

where: T – temperature; K – damage rate; E_m^i – migration energy of interstitials; N_0 – constant, containing the necessary geometrical factors, including cross section of interstitials loss on the fixed sinks, existing in material, such as grain boundaries, dislocations lines etc.

Really received in work [7] data about proportionality of interstitial loops number density to a square root from damage rate irrespective of a type of irradiation (fast neutrons, D-T neutrons, 1 MeV electrons) speak that nucleation of interstitial loops at an irradiation temperature more than 400°C occurs at the expense of reactions between free interstitials, and not as the result of cascades slams, i.e. it is possible to assume, that the cascade damages do not play the main role in nucleation of interstitial loops in Fe-Cr-Ni alloys at irradiation temperatures 400...500°C.

It is well-known that subsequent formation of dislocation network in irradiated annealed austenitic stainless steels is enough complicated process which has few stages:

- formation from interstitial clusters fault (Frank) loops with b=a/3<111>ii on close-packed planes (111) in FCC lattice of stainless steels;
- transformation of Frank loops to perfect loops due to faultness reaction with Shockley loops:

$$a/3 < 111 >+ a/6 < 112 >= a/2 < 110 > i$$

prismatic loops with b=a/2 < 110 > i

tion in different planes and three dimensions dislocations network formation.

The dependence of irradiation rate on dislocation structure parameters are summarized in table 1 [7].

 Table 1

 Dislocation structure data with different irradiation

 intensity

| Intensity | | | | | |
|---|--------------------------------------|---|--|--|--|
| Defect structure | Dependence on irradiation rate | Consequence of accelerated irradi- ation | | | |
| Number density of interstitial loops | $\Phi^{\scriptscriptstyle 1/2}$ | enhanced | | | |
| Loop size | $\Phi^{-1/2}$ | suppressed | | | |
| Dislocation density | no depen- dence | unchanged | | | |
| Number of intersti- tials in interstitial loops | $\Phi^{-1/2}$ | suppressed | | | |

The enhancement and suppression are towards opposite directions between the number density of dislocation loops and the total area of dislocation loops, while the dislocation density is kept comparatively unchanged.

Dislocation structure and void swelling at model austenitic alloys. In model austenitic alloys only these components show sensitivity to dpa rate. Formation of network is critical point for voids formation, because well known that without critical number density of dislocations voids can not be formed [2].

Data about features of dislocations behaviour under different dpa rates (especially for neutron experiments) are enough limited, especially these study does not provide strong clues illuminating the microstructural origins of the sensitivity of swelling to dpa rate.

These limited results confirm that the earliest and most sensitive component of microstructure to both temperature and especially displacement rate was found to be the Frank loops, whose rate of unfaulting determines when the dislocation network starts to evolve. The second most sensitive component was found to be the void microstructure, which co-evolves with the loop and dislocation microstructure.

A lower dose enhances network dislocation formation. This is caused by enhanced loop growth and unfaulting. (Mutual recombination rate of interstitial and vacancy is thought to be much larger). Enhanced loop growth with lower density is observed at lower dose rates, resulting in earlier loop unfaulting and enhanced rate of network dislocation formation (fig. 4) [8].



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Fig. 4. Effect of dose rate on dislocation density during neutron irradiation at 426°C in Fe-15Cr-16Ni alloy [8]

The strong onset of unfaulting and dislocation evolution at very low dpa rates erases any meaningful evidence of the dependence of Frank loop nucleation, but the charged particle experiment mentioned early clear shows that Frank loop nucleation is strongly sensitive to dpa rate.

Muroga et al [9] also showed that saturation density of Frank interstitial loops increased with (dpa rate)^{1/2} over more than three orders of magnitude in dpa rateloop density of Fe-Cr-Ni austenitic ternaries irradiated with fast neutrons, fusion neutrons and electrons at around 723 K is nearly proportional to the square root of nominal damage rate.

In general higher densities of loops develop concurrently with smaller loop sizes and therefore different probabilities of intersection, interaction and unfaulting. Network formation is proceeded by unfaulting and interaction of dislocation loops, whose size and density are very flux – sensitive.

At lower dose rates loops are low in density and much larger in size. This yields earlier unfaulting of loops and earlier formation of a dislocation network. It is meaning that loops at smaller sizes (during high dpa rate irradiation) are more stable against interaction and self – unfaulting, in this case network formation is restricted.

Presented data demonstrate that in austenitic stainless steels duration of transient regime of swelling directly correlates well with dose required to form a dislocation network.

In work [8] dislocation evolution model based on dose rate effects was suggested. The process microstructurally involves irradiation – assisted climb and glide of network components, annihilation of dislocation of opposite sign, and the nucleation, growth and unfaulting of Frank interstitial loops to form new network.

Results of calculations schematically summarized in fig. 5. These results may be useful for more complicated commercial alloys in cold-worked condition under modification of a part, which defined strength of obstacles of climbing motion.

Fig. 5. Dislocation evolution model [8]. At lower dose rates network formation by loop unfaulting occurs at much lower doses

At lower dose rate, enhanced dislocation evolution increase swelling by shortening the incubation dose for the onset of steady state swelling. Accelerated nucleation of dislocation loops enhances voids nucleation at low dose rates. Nucleated low density of voids can grow larger, because high density of dislocation sinks provides enough vacancy super saturation for these voids. (fig. 6). The balance is considered to increase swelling rapidly from very low dose region. Dose rate dependence of the net vacancy flux to void can be described a such equation:

$$dr/r=1/r(Z_{vv}D_{vv}C_{v}-Z_{vi}D_{i}C_{i}), \qquad (2)$$

where r - void radius, $D_{i,v} - diffusion$ coefficients of interstitials and vacancies, $C_{i,v} - point$ defects concentration, Zvi, Zvv - bias factor of voids for interstitials and vacancies.



Fig. 6. Trends of dpa rate effect on microstructural evolution of Fe-15Cr-16Ni alloy [8]

The net vacancy flux was found to be proportional to $(dpa/sec)^{1/2}$, indicating mutual recombination dominates point defect annihilation even though the point defect sink population is well developed.

Dislocation structure in commercial stainless steels. In table 2 are presented microstructural data from few commercial stainless steels irradiated with different dpa rate. Seems that dpa rate influence on all components of steels.

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| Steel | D, dpa | T,°C | Dpa rate, dpa/sec | $\rho_{\text{Fr.l}}, \text{cm}^{-3}$ | $\rho_{\text{Tot,}}, cm^{-2}$ | ρ_{voids}, cm^{-3} | $\Delta V/V$, % | L _{pr} , nm | ρ_{pr}, cm^{-3} | Ref |
|---------|--------|------|----------------------|--------------------------------------|-------------------------------|-------------------------|------------------|----------------------|----------------------|------|
| 304 | 3 | 392 | 7.9×10 ⁻⁷ | 1.45×10 ¹⁶ | 8×10 ¹⁰ | 4×10 ¹⁵ | 0.03 | 18 | 1×10 ¹⁴ | [10] |
| 304 | 2.6 | 371 | 0.8×10 ⁻⁷ | 7.0×10 ¹⁵ | 3.5×10 ¹⁰ | 7×10 ¹⁴ | 0.005 | 9 | 2×10 ¹⁴ | |
| 316 | 3.3 | 392 | 8.4×10 ⁻⁷ | 8×10 ¹⁴ | 3.2×10 ¹⁰ | 5×10 ¹² | 0.02 | 40 | 1.1×10 ¹⁵ | |
| 316 | 3.3 | 371 | 1×10 ⁻⁷ | 2.1×10 ¹⁵ | 3.1×10 ¹⁰ | 4.3×10 ¹⁴ | 0.01 | 17 | 1.2×10 ¹⁴ | |
| 316+ Ti | 50 | 675 | 5×10 ⁻⁴ | | 1.8×10 ¹⁰ | | | | 6.2×10 ¹⁴ | [11] |
| 316+ Ti | 32 | 555 | 5×10-7 | | 3.1×10 ¹⁰ | 2×10 ¹⁴ | | | 3×10 ¹⁵ | |
| EI-847 | 2 | 650 | l×10-3 | 2.2×10 ¹⁵ | | 6×10 ¹⁴ | | | 2×10 ¹⁴ | [12] |

Some microstructural data in few commercial steels under irradiation with different dpa rate

Azam [13] demonstrated linear increasing of total dislocation density in AISI 316 in variations of dpa rate from 2×10^{-3} dpa/sec up to 13×10^{-3} dpa/sec (T_{in}=600°C).

LeNaour [14] results directly showed distinctive difference in dislocation behaviour in solution annealed 316 at different dose rates:

- 4×10^{-7} dpa/s no loops observed, increasing dislocation density to almost saturation value results essentially from dislocation climb (sources, helix formation, dipoles) and from the emission of dislocations at precipitate interface. Heterogeneous distribution of dislocations being maximum in the vicinity of original dislocations and precipitation. Saturation of dislocation density is around 20 dpa at comparatively low level 5...6×10¹³ cm⁻², at this dose dislocation network tends to become uniform.
- 13.3×10^{-7} dpa/s the increase of dislocation density is mainly due to the nucleation and growth of interstitial loops. These loops are observed throughout the dose range investigated, their number being almost constant (5×10⁻¹³ cm⁻²). The saturation of the total dislocation density seems to occur at a comparatively higher dose (~36dpa).

Enhanced loop evolution drives the total dislocation density to saturate at a level five times higher than that found at the low dose rate. Also the higher dose rate prolongs loop evolution ceases after 11 dpa at low dose rate.

Experimental results pronounce some features in dpa rate influence on dislocation structure in irradiated steels in annealed (SA) and cold-worked (CW) conditions and in steels which have distinctions in chemical composition.

Behaviour of dislocation structure in technologically important cold worked steels with high degree of deformation (15...30%) is ever more complicated in comparison with SA steels. Original high dislocation density can essentially modify dislocation structure evolution in irradiated steels.

For stainless steels the neutron-induced saturation density of network dislocations in near $(6\pm3)\times10^{10}$ cm⁻², in dependent from many factors, particular irradiation temperature and displacement rate. Typical tendency for the dislocation microstructure to evolve toward a saturation or quasi-steady state level [1] case involves an or-

der of magnitude reduction in the dislocation density of cold worked steels and a comparable or large increase in the density of annealed steels.

The decrease in swelling when dose rate increases is essentially due to a nucleation effect. Two main factors could play a role on dose rate effect:

- at the higher dose rate loop nucleation occurs because a higher initial supersaturation leading to a rapid increase of dislocation density to a level two times higher than at 4×10^{-7} dpa/sec. This minimizes the vacancy peak in the vicinity of preexisting dislocations.
- since the number of voids is smaller than at 4×10⁻⁷dpa/sec, nickel depletion is minimized, that is why void sizes at higher doses are smaller at high dose rate.

Segregation and precipitation. Microchemistry only few data exist about dpa rate on level of segregation processes in austenitic stainless steels.

Results of work [16] showed that as the displacement rate decreases the amount of nickel enrichment and chromium depletion in SA 304 increases on grain boundaries and on precipitates interface (fig. 7,a). The greater segregation in the EBR-II materials is likely attributable to a dose rate difference, ever at comparatively low irradiation temperatures (375...400°C).

Calculations by rate theory for ratio of recombination point defects fraction to fraction of point defects disappearing on sinks: at relevant temperature decreasing of dpa rate leads to decreasing recombination and increasing the segregation on sinks (fig. 7,b).

The matrix solute depletion affects microstructural evolution in two ways: by changing the defect diffusion coefficients and by altering the dislocation bias. Le Naour data [14] showed that for the two investigated dose rates there is an important segregation of Ni at voids. The nickel content of the matrix is minimum between two voids and increases when the distance to the void decreases. It was shown that these segregation data are slightly dose rate dependent, the nickel segregation increases with dose rate increasing (maximum value of nickel concentration near voids for a sample irradiated at 13×10^{-7} dpa/s and 20.5 dpa achieve 34%, at the same time at 4×10^{-7} dpa/s and 21 dpa nickel level consists 29%).

Nickel depletion of the matrix due to precipitation (detectable only at 20 dpa) always remains much smaller than the depletion due to segregation at voids, but, of course it is depends from distribution as precipitates as well voids.



Fig. 7. Radiation-induced segregation in samples with different dpa rate: a) grain-boundary segregation [16], b) authors theoretical calculations [24]

Precipitates that develop in austenitic stainless steels type 316 during irradiation play a dominant role in the alloy response.

The role of second phase precipitates on swelling behaviour appears to arise primarily from their influence on the distribution of minor elements and the resultant changes in matrix composition. Beside it nature of the precipitate-matrix interface may influence swelling due to increasing recombination of point defects.

The dependence of various precipitation sequences on displacement rate (nickel silicides, which are result of RIS) or on time at temperature (thermally stable phases) is very important in the rate of microchemical evolution of the alloy matrix [17].

Most phases found in alloys based on the Fe-Cr-Ni system are metastable and are very sensitive to both thermal and radiation conditions.

Precipitation behaviour seems to be dose rate dependent. For instance Laves phase is abundant at the lower dose rate, its volume fraction decreases progressively with increasing dose rate until it disappears for a dose rate of ~ 14×10^{-7} dpa/sec. Obviously the exists dose rate beyond which no Laves phase is present. Thus Laves phase has been detected at 600°C in SA 316 irradiated in a wide dose range at a flux of 4...7.5×10⁻⁷ dpa/s and is absent in specimens irradiated at higher dose rate 13 $\times 10^{-7}$ dpa/s [14]. Really in own experiments of author of this report on such steels irradiated with heavy ions (dpa rate $\sim 10^{-3}$ dpa/s) we never saw [18] formation of Laves phase in such type steels.

Gilbon et al [19] demonstrate that the dose rate is a very important factor for the evolution of precipitates as for the chemical composition of precipitates. He showed that when dose rate varies from 10.94×10^{-7} dpa/sec down to 3.8×10^{-7} dpa/sec (irradiation temperature ~590... 595°C), the following changes in microstructure are ascertained:

- 1. Drastical decreasing of Laves phases number density from 20×10^{12} to 5×10^{12} cm⁻³. For the lower dose rate these precipitates lie almost entirely near twin boundaries. The measured lattice parameters are much closer to those of standard Fe₂Mo. Nickel content of these precipitates has decreased from 18 to 11%. This result is in agreement with the fact that the Laves phases are generally considered as irradiation enhanced and modified.
- 2. The density of Ni₃Ti platelets is decreased from 3.7×10^{14} to 1.3×10^{14} cm⁻³ and their diameter is also slightly reduced.
- 3. The density of TiC, which would normally appear by thermal annealing, increased from less than 4×10^{15} to nearly 10^{16} cm⁻³, when dose rate decreases. At the lowest dose rate, the TiC particles are the main intragranular precipitates and they are coincident with the cellular array of dislocation network.
- 4. For the dose rate of 3.8×10^{-7} dpa/sec some big carbides of the M₂₃C₆ type are found at grain boundaries.

As a result the amount of precipitate at a given fluence but different flux (different dpa rate) is greatest for low flux irradiation. Whereas one might expect more precipitates to form in those specimens exposed to the highest fluence, the lower fluence specimens which spent a longer time at reactor at lower flux.

It has been reported that the fine distribution of MC particles causes swelling suppression by offering recombination centers for vacancies and interstitials. It is well known that precipitation behaviour during irradiation is determined by a balance between precipitate shrinkage due to irradiation resolution and re-precipitation accelerated by irradiation-enhanced diffusion. Some results [12] about the effect of dose rate on the precipitation behaviour indicate that the balance between them is sensitive to the dose rate in the case of the fine TiC precipitates. It seems that the reduction in the amount of TiC precipitation with increase in dose rate can be caused by the enhancement of precipitate shrinkage due to irradiation resolution and the shorter irradiation time.

It is impression that in stainless steels exists competition between radiation-induced phases (such as $-\gamma$ and G-phase) and thermally activated (time dependent) carbides and intermetallic phases (fig. 8).

It was shown that decreasing of dpa rate leads to more intensive precipitation of Ni and Si-content phases

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(γ ' and G-phase). As a result Ni and Si content at matrix decreases and create possibilities for intensive swelling.

Experimental results show that features of such competition are such:

- radiation-induced phases γ(Ni₃Si) and G-phases (Ti₆Ni₁₆Si₇), which are formed mainly due to high diffusion mobility of nickel and silicon), are more stable at high dose rate and lower irradiation temperature. Stability of these phases is defined by mechanisms of RID and mainly RIS;
- slowly formed carbide and intermetallic phases can not survive at high dose rates. These phases can be more stable at higher irradiation temperature, where rates of diffusion are higher.



Fig. 8. Influence of dpa rate on precipitates stability: $a - \gamma'$ -phase (Ni₃Si) formation in alloys Fe-15Cr-20Ni-Si at different dpa rate ~ 10⁻⁶ dpa/s in EBR-II, 2×10³dpa/s for ions [1]; b - G-phase (Ti₆Ni₁₆Si₇) stability during irradiation in alloys Fe-18Cr-10Ni-Ti

at ~10⁶ dpa/s in BOR-60 and 2×10^{-2} dpa/s for ions [24]

It is exists critical combination between T and dpa rate at which stability of any precipitates must increase at low dpa rate at higher temperatures, or long time and high temperatures and formation of such precipitates can change matrix composition. If flux-time effect on precipitates stability is enough strong in narrow temperature interval, samples with low dose rate can transform to region, where transition period is dramatically short.

Theoretical description shows that decreasing of dpa rate must leads to increasing of dose at which the same level of segregation can be achieved [20]. It is known that resistance to void swelling in iron-nickel-chromium alloys changes with bulk vacancy diffusivity and the degree of major element segregation. Enrichment of nickel and depletion of chromium reduces the vacancy flux to voids, decreasing the void nucleation rate and limiting the magnitude of void growth.

Voids and swelling in commercial steels. The most of experimental results show the same tendency as for model alloys – higher swelling with lower dpa rate. Tenbrink [21] found that swelling to increase distinctly with decreasing displacement rate. This behaviour can be explained enhanced nucleation rate of small voids at the higher displacement rates leading to a lower growth rate of the large voids.

In SA 316 maximum void nucleation rate (2×10^{12}) voids per dpa) occurs between 12 and 20 dpa and almost saturates at the onset of steady state swelling, whereas the mean void size begins only to increase when the steady state is established. Comparison of results at close doses and temperatures shows that swelling is higher at low dose rate (4×10^{-7}) compare to 13.33 $\times10^{-7}$ dpa/sec) mainly due to higher number density [14].

Which physical mechanisms are responsible for such structure evolution and corresponding swelling behaviour under different dose rate?

Absence of visible radiation damage in microstructure of SA 304 which was irradiated at low dose rate at 600°C allow to say that under such irradiation conditions practically all defects recombine. This could be due to the existence of defect-trapping impurity clouds in the vicinity of preexisting dislocations which would locally greatly enhance recombination, thus impeding dislocation climb and the required conditions for void formation. The incubation period would the correspond to the time necessary to form a given amount of free dislocations.

It is also important to note that although dose rate does not seem to affect the same component of the microstructure in SA and CW 316-respectively the dislocations and precipitates a common factor seems to appear. At low doses voids are seen in both cases to nucleate in the vicinity of the component affected by dose rate (dislocations in SA 316, Laves phase in CW 316), whereas at the highest dose rate void are almost uniformly distributed in the swollen regions and not clearly associated with any component of the microstructure. This could explain why the same trends are observed for the effect of dose rate on the swelling of both materials.

In CW 316 at low dose rate $1.5...4 \times 10^{-7}$ dpa/s voids are only associated with Laves phase precipitates, their number density is relatively small ~4×10¹¹...10¹² cm⁻³ At intermediate dose rate ~7.5×10⁻⁷ dpa/s double distribution of voids is observed: ~7×10¹² cm⁻³ large voids associated with Laves phase and almost uniform distribution of 5×10¹³ cm⁻³ voids of ~5nm: at this dose rate all the swelling is due to associated voids. Increasing of dose rate up to 12×10⁻⁷ leads to uniform distribution of voids in the specimen and formation different voids populations-large voids ~150nm associated more probably with Laves phase, smaller voids ~70 nm are clearly not associated with any precipitates and largely contribute to the overall swelling and as at 7.5×10⁻⁷ dpa/s a population of 5×10¹³ cm⁻³ voids of size near 15 nm. Such features can lead to some assumptions on the behaviour of CW 316 irradiated at 600°C.

Intermetallic phases (particular Laves phase) being a heterogeneous nucleation site for voids and being essentially observed in the lowest dose rate range. It is naturally that at low dose rate the incubation dose for swelling is reduced.

In the highest dose rate range the end of incubation dose is attained through the uniform nucleation of a great number of voids; the segregation of nickel on voids can change the void bias and enhance swelling rate. In CW 316 and in SA 316 the onset of the steady state swelling regime seems correlated with the development in the specimens of a new irradiation induced dislocation network.

Explanation of such difference in the void population can be connected with the precipitates behaviour. Where Laves phase precipitation is intense void nucleation is heterogeneous and those voids associated with Laves particles account alone for the overall swelling. On the contrary in the highest dose range where no Laves phase is present most of the swelling is accounted for by a uniform distribution of unassociated voids; the segregation effects in this case deplete the matrix in nickel.

TEM observations [14] show that the in incubation period of the high temperature swelling is closely associated with the setting up of an equilibrium micro structure the nature and the history of which depend on the damage rate for a given temperature. This equilibrium microstructure results from a competition between two types of possible evolution:

- the nucleation and growth of dislocation loops at high damage rates;
- the climb of the initial dislocation network at low damage rate. When climb occurs, there is also a precipitation of M₂₃C₆, and one may think that the dislocations have been cleaned of their impurity clouds which inhibited swelling. On the other hand if the microstructure takes the other route, the loops; there is no swelling.

It seems that dpa rate effects can be sensitive to irradiation temperature and structure state of irradiated steels.

Cole et al [22] reported data on comparing of swelling in hexagonal ducts which were irradiated at different dose-rates, but each pair was irradiated at similar temperature and dose. The difference in dose-rate is about a factor of two for each of these pairs. Both the calculated void swelling and measured bulk swelling are greater at the lower dose-rate for all pairs of samples. Void diameters were also consistently greater at the lower dose-rate, while the overall void density varied. The data show that the increases in void diameter and bulk swelling due to decreasing dose-rate were seen at all temperatures and doses examined. T. Allen et al [23] showed that even no significant effect of dose rate is evident in their study, the samples irradiated at a lower dose rate do show greater swelling .Plots the swelling for six different pairs of samples chose to have similar irradiation temperature and dose, but differing in dose

rate by a factor of two demonstrate that in each case, the sample irradiated with lower rate shows greater swelling.

EFFECTS OF STRESS

In active zones of nuclear reactors the majority of structural elements, including claddings and wrappers, works under the stress, under the stress influence is a damage layer during ion or electron irradiation. It was suggested that stress condition can stimulate swelling development as result of interaction with defects in irradiated materials through their "own" fields of stresses. Level of stresses that are expected to cause enhanced swelling on the fuel claddings (residual stress due to final cold working of cladding and gas pressure in fuel pin due to fission products gas release) under exploitation was estimated at about 50-200 MPa. The most of experimental data on investigated austenitic steels are received for such stress level.

Unfortunately the possible role of stress on swelling behaviour was discussed for some time as various contradictory data sets were complied and many previous results and explanation were partially revised [1].

The effect of stress-induced swelling has been found in many alloys at different irradiation temperatures and thermo mechanical treatments [25].Practically in all cases stress increases void swelling in irradiated austenitic stainless steels (fig. 9) [26].



Fig. 9. Effect of stress on swelling of two modified 316 SS irradiated in the form of pressurized tubes in PHENIX [26]

Now there are few key problems for understanding of the stress role in swelling behaviour and extrapolation of its influence up to higher doses:

- 1) Indicate whether the stress affects the incubation period, the steady-state swelling or both;
- 2) Is the effect of stress on swelling operates on both annealed and cold-worked steels?
- 3) Which type of stress (hoop or tensile) is more pronounced?
- 4) Which temperature interval stress influence is more effective?

Really up to now two different approaches are usually used to describe the effect of stress on swelling. The stress is assumed either to reduce the incubation dose or to increase the steady state swelling rate [27]. It is important to distinguish between these two aspects of the

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swelling phenomena in order to extrapolate its behaviour to higher fluences with the great accuracy. Extrapolation will be different for a growth-affected process than for a nucleation – affected processes.

On the one hand the stress can enhance the void growth. This would lead to bigger voids in a stressed sample compared to an unstressed one. On the other hand the stress can accelerate the void nucleation. This would lead to a higher void concentration. The reduction of the incubation dose corresponds to accelerated void nucleation and will lead to a higher void concentration. The increase steady state swelling rate is related to a bigger average void diameter. Both effects have already been found in enough wide temperature interval (400... $600 \in C$) of irradiation [28].

The analysis of the stress dependence of the stress induced swelling yields no big difference between the mechanisms of reduced incubation dose and increased steady state swelling rate in the steel 1.4981 [29]. Mathematically these effects can be described as follows: after the swelling rate reached the steady state swelling rate:

$$\Delta V/Vo(\sigma_{hy}) = \Delta V/Vo(\sigma_{hy}=0) \times (1 + A \times \sigma_{hy}), \quad (3)$$

$$\Delta V/Vo(\sigma_{hy}) = \Delta V/Vo(\sigma_{hy}=0) + B \times \sigma_{hy}, \qquad (4)$$

where $\Delta V/Vo$ – stress free volume change; σ_{hy} – hydrostatic stress; $\sigma_h = l/3(\sigma_1 + \sigma_2 + \sigma_3) = l/2\sigma_{\theta}$; $\sigma_{1,2,3}$ – axial; σ_{θ} – shear stresses A and B – material constants

The values found for the constants A and B are in the same order of magnitude as reported for other austenitic steels [27]. The average values of the constants in the dose range 50...105 dpa (420°C) are: $A - 4 \pm 2...9 \pm 1.5 \times 10^{-3} MPa^{-1}$, $B - 1.5 \pm 2...3.5 \pm 2 \times 10^{-4} MPa^{-1}$.

The results of the TEM-analysis on average void diameter and void concentration support that the effect of stress on the void structure is rather complicated (table 3).

 Table 3

 Average void diameter and void concentration in

 DIN 1.4970 steel [27]. (T_{irr}=420°C D=106 dpa NRT)

 [27]

| Stress, MPa | Voids diame- ter, nm | Voids concen- tration, $\times 10^{15}$ cm ⁻³ | ΔV/V,% | | |
|-------------|-------------------------|--|--------|--|--|
| 0 | 25 | 3.8 | 4.4 | | |
| 60 | 27 | 3.2 | 4.9 | | |
| 120 | 27 | 3.7 | 5.6 | | |

Due to distribution of big voids it was assumed the accelerated void growth as the cause for stress-induced swelling, but the accelerates void nucleation can be excluded. At the same time Russian data [30] directly showed that applied stress (up to 200 MPa) leads to increasing of void swelling due to void number density, where this effect on void growth is very low (fig. 10).



Fig. 10. Microstructural data obtained by microscopy of 16Crl5Ni3MoNb (EI-847) specimens irradiated at 480°C to 60 dpa [30]

Applied stress influence the void number density, leading to its increasing, but not the void size or dislocation density (fig. 10). Value of the constant for these steels was estimated as $A=7.98 \times 10^{3} MPa^{-1}$.

Results on stress-affected swelling published earlier by Bates and Gilbert [31] were confirmed later by Shamardin and coworkers [32].

Void swelling at given dose and temperature usually increases linearly with the applied stress until the yield stress is exceeded, and then the swelling drops. The net effect of stress is only exerted on the incubation dose, and does not on the steady-state swelling rate. The results of TEM measurements for voidage parameters in irradiated steel 16Cr15Ni3MoNb showed that with increasing stress level (in the range 0...150 MPa) void number density increasing has place with a simultaneous decrease of the voids volume (particular, for SA condition) (fig. 11), according to TEM data the maximum swelling is noted in this case at a stress of 120 MPa and furthermore, the stress increase leads to a swelling decline of from 1.2% at 120 MPa to 0.8% at 150 MPa. Then, at 150 MPa, number density increasing does not compensate the average volume decreasing and results in a swelling decline.

Direct comparison of stress influence on swelling in SA and CW steels of the same heats are very limited, cold-worked material can responds differently from annealed material, and often in unexpected way, depending upon fabrication history and stress state.

The behaviour of voidage parameters in steel 16Crl5Ni3MoNb in cold worked condition follow the same tendency for this stress levels – increasing the number density and decreasing of void volume fraction [32].

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Experimental results of work [33] about stress influence on swelling of pure AISI 316 steel also can be summarized in this way:

- external stresses (0-100MPa) reduce the incubation period for void formation;
- the void growth rate is unaltered by external stresses.



Fig. 11. Changes in the irradiation voidage parameters of 16Crl5Ni3MoNb steel (SA version) versus stress in fuel pin claddings [32]

Next important summary of this work was: both, incubation period and growth rate are indifferent upon reversion of stress from uniaxial to compression. The similar confirmed results were received by T. Lauritzen et al [34] which supported that the magnitude of swelling is independent of the sign of the applied stress) (fig. 12) and they suggested that the mechanism for stress-enhanced swelling in this (AISI 316) steel may be a sensitive function of the magnitude of the applied stress.



Fig. 12. Void swelling in 20% cold worked AISI 316 as a function of stress state and fluence [34]

Efficiency of stress influence in a great degree depends from irradiation temperature, but there are some uncertainties in temperature influence. Most stress-affected swelling positions postulated that stress could only affect swelling at relatively high temperature (fig. 13); the low temperature regime exhibits a stress effect with little or no temperature dependence. It appears, however, that stress enhances swelling at all temperatures, but it more sluggish in exerting its influence at lower irradiation temperature [35].



Fig. 13. Stress enhanced density changes observed in 20% cold worked AISI 316 [35]

Really Walters and Flinn data [36] on swelling in annealed 304 SS irradiated at temperature to 380°C to 1.4 and 5.1410²²n/cm² (E>0.1MeV) in both the unstressed and stressed conditions (σ = 188 MPa) found no significant dependence of irradiation-induced swelling on applied stress. It can explained that stress effect on void density and volume exhibits a strong temperature dependence. From author's point of view swelling level at these temperatures is too small for representative understanding of stress influence.

The most of experiments are performed at intermediate temperature range typical for fast reactors: 450... 550°C and at this area stress effect is well determined-it accelerate voids formation and stimulate their development [26].

Stress provoke changes in swelling value and development by the influence on processes of generation and diffusion of point defects, formation of dislocation network and microchemical changes, i.m., practically on all components of defect structure. The microstructural development is the accelerated by the applied tensile stress.

Point defects-calculations showed [37] that rate of generation of point defects and their diffusion mobility (as possible result of reducing the migration energy) are increased under influence of tension stress.

Dislocation structure and voids – next step of microstructure development in stressed material are pronounced with such features.

First of all it is important to note that most stress effects studies are conducted at stress levels below the yield strength, such the network dislocations and dislo-

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cation loops interact primarily via their competition for point defects.

Porollo [38] and Loguntsev [39] paid attention on some aspects in dislocation evolution of stressed steels which may cast some light on stress affected -swelling data of Bates and Gilbert [31] and Shamardin with coworkers [32] – they found that if the stress is below the yield stress Frank loops are formed during irradiation. Above this stress, however, Frank loops are not formed and dislocation networks and cells of varying perfection form instead. Since both dislocation loops and network dislocations are normally cited to influence the nucleation of voids, it is not surprising that the absence of Frank loops at stress level above the yield somewhat delay the onset of accelerated swelling.

The primary effect of stress is to enhance the nucleation of voids and Frank loops. A secondary effect involves stress-enhanced growth of the loops. It was suggested [40] that the shear components of stress operates primarily on the void number density and that that Frank loops evolution and loop-associated precipitates may be responsible for the accelerated nucleation of voids.

Number density of voids should be related to stress in the following manner

$$\rho_{\rm V}(\sigma_{\rm H})/\rho_{\rm V}(0) \approx \exp(n\Omega\sigma_{\rm H}/kT),$$
 (5)

where Ω is atomic volume, $\sigma_{\rm H}$ -hydrostatic pressure. Total loop density can be written as follow:

 $\rho(\sigma)_{\text{Tot}} \approx \rho_{\text{Tot}}(0) \exp(\pi R_0 2b/kT) \sigma_{\text{H}}, \qquad (6)$

where R - loop size, b-strength of Burgers vector.

Comparison of these two equations indicates that both the void and the loop population evolve with a similar response to the applied stress rate.

The Igata observations [41] also suggest the possibility of void nucleation as a direct consequence of the presence of a dislocation loop. The mechanism controlling of stress dependence of the incubation dose of swelling was interpreted as result of stress-affected dislocations behaviour. Due to high local strain field near the dislocation loops separated absorption of interstitials at the peripheral sites of dislocation loops and vacancies at the center of dislocation loops.

Nucleation of dislocation loops will be defined as competition of two different mechanisms (enhanced nucleation with applied stress and suppress by preexisting dislocations due to their absorption of interstitials):

$$N=N_0 \exp(n\sigma b^3/kT)[1-\gamma \pi R^2(\sigma-\sigma_I)^2/(\alpha \mu b^2)], \quad (7)$$

where dislocation density ρ is shown as $(\sigma - \sigma_1)^2/(\alpha \mu b^2)$; N₀ – loop number density under stress free condition; σ – applied stress; R – -dislocation capture radius for interstitials; n – critical number of atoms for loop nucleation; γ – capture efficiency; σ_I – frictional stress; μ – shear modulus; b – strength of Burgers vector and α – coefficient for hardening by dislocation.

The degree to which the dislocation loops and the dislocation network which evolve from the loops are effected by the stress is dependent upon the angular relationship between the stress field and the crystal orientation. Frank loops are found to have higher densities on close packed planes {111} having the highest normal stresses. Anisotropy distribution of dislocation Frank loops evidence action in material SIPA (Stress Induced Preferential Absorption) mechanism which can stimulate loops nucleation.

Another working mechanism (PAG) stress- induced climb and glide under the stress. In materials, which are irradiated under the stress, dislocations are really more mobile and network formation is going faster, dislocation density achieves the saturation level faster. This mechanism would tend to accelerate swelling by shortening the incubation period and also exhibits a weak dependence on temperature. This mechanisms seems important only in SA steels, in CW material the pre-irradiation dislocation microstructure exists at levels comparable to that produced by radiation [40].

Mechanisms of stress influence on development of void swelling through influence on accompany processes add to mechanisms which are based on changing energetic characteristics of nucleated voids and their thermal stability (SANV – Stress-Assisted Void Nucleation) mechanism [42]. At this mechanism it supported that efficiency of voids as sinks for point defects Z_{vj} depends from level of external stresses, voids possess bias factor to interstitials, and bias level depends from stresses es level and decreases with increasing of voids size

$$Z_{\nu j} = 1 = \left[\frac{(1-\nu)^2}{36\pi(1-\nu)} \times \frac{G\Delta V_j^2}{kT}\right]^{1/3} \frac{1}{r_{\nu}} - \frac{3}{56} \frac{\alpha_j^G}{G^2} \left(\frac{2\gamma}{r_{\nu}} - \sigma_n\right)^2$$
(8)

where ν – Poisson's coefficient; ΔV_j – change of volume, connected with formation of point defect type j; r_{ν} – voids radius; γ – surface energy; G – shear modulus; α_i^G – shift polarization of point defects.

Efficiency of interstitials interaction with voids decreases stronger, because tensile stress decreases efficiency of interaction of voids with point defects, that's why voids bias to interstitials decreases and barrier for voids nucleation is minimized.

All mechanism described above are enough efficient at temperatures below temperature of maximum swelling.

It was suggested earlier that the effect of stress can be more pronounced at high temperatures of irradiation.

Emission of point defects from voids and influence of this process can essentially influence swelling at high irradiation temperatures [43]. Thermally equilibrium vacancy concentration near the voids surface is described as:

$$C_V^V = C_V^0 \exp\left[\left(\frac{2\gamma}{r_V} - PG - \sigma_n\right)\frac{\Omega}{kT}\right],\qquad(9)$$

where γ – surface energy; σ_n – hydrostatic pressure; Ω – atomic volume.

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These equations demonstrate that in field of tension stress rate of thermal emission of vacancies from voids decreases. It leads to increasing of thermal stability of voids and stimulate their development.

Segregation and precipitation. This sensitivity has been shown to be related to the stress sensitivity of the rate of formation of intermetallic phases at relatively high temperatures. Below 700°C the intermetallic contribution is expected to be small. The low temperature radiation-induced phases and carbosilicides that form at low temperatures seems to be insensitive to applied stress. This has been verified experimentally for the γ' phase in AISI 316 and has been inferred for the G-phase from the relative stress – insensitivity of swelling of annealed AISI 316 at low temperatures [36].

Experimental data about stress influence on irradiation-induced microchemical changes (segregation and precipitation) are enough limited, but some data directly showed precipitation increasing as stress results (table 4)

Table 4 Parameters of second phases in 16Crl5Ni3MoNb steel (T_{irr}=450°C, fluence 6.2×10²⁶n/m², E>0.1 MeV) at different stresses [32]

| Cladding Stress, MPa | Average Particle Size, 10 ⁻⁸ cm | Volume ra- tio of parti- cles, % | Particle Density, 10 ¹⁵ cm ⁻³ | Average in- ter-particles distance, ×10 ⁻⁸ cm |
|----------------------------|---|---|---|---|
| 90 | 75 | 0.16 | 2.2 | 1360 |
| 120 | 85 | 0.33 | 4.7 | 1070 |

It was shown that application of stress accelerates segregation processes in AISI 316 and leads to the radiation-induced nickel removal process. [44]. This effect is negligible at 400°C (fig. 14), but it is obvious at 550°C (fig. 15).

These results correlate well with the discussed temperature dependence of radiation-induced segregation [17].

Stress does not alter significantly the mechanism of swelling or the rates of solute segregation, but merely allows swelling to start at lower dose levels. A plausible explanation for the stress effect is that the degree of microchemical segregation required for swelling to commence is merely decreased by the application of a volumetric stress or the associated application of strain energy to the system. Such an effect may be manifest in a reduction in the effective stacking fault energy for dislocation loops or increases in defect mobilities. There are some additional stress – affected mechanisms which can influence swelling.

Akasaka et coworkers [45] paid attention on formation of secondary stress, which was occurred by non uniform void structure in cladding enhances swelling due to temperature gradient and which can influenced on increasing of swelling level. Some experimental evidence from Dounreay Fast Reactor that swelling in fuel pin cladding and unstressed samples of cold-worked 316 is much the same after a dose about 26 dpa. These results support idea that stress may have only effects at high irradiation temperature [46].

The swelling data sets for fuel pin cladding and non fueled cladding of the few lots irradiated at First FFTF Core exhibits no gross discrepancies. Both sets follow a bilinear relationship and do not deviate noticeably from a linear-after-incubation behaviour. These results lend confidence to the usage of non fueled cladding data to predict the swelling of fast reactor fuel pins [47].



Fig. 14. Comparison of nickel content of precipitates in heat-treat and cold-worked AISI 316 at 400°C [44]



Fig. 15. Effect of stress on nickel segregation into precipitates in 10% cold-worked AISI 316 at 550°C [44]

Majority of the results on AISI 316 [1] imply that the stress acts mainly by reducing the incubation dose instead by increasing the void-growth rate. It was suggested that, once, a stress had exerted its influence on swelling by reducing the incubation period, a subsequent variation of the stress level has no further influence on the steady state void growth. Only for AISI 316 there seem to be enough data to warrant the conclusion that is the dominant physical mechanism for swelling enhancing.

TEMPERATURE HISTORY

It is well known that small details of reactor operation that ensure safe, reliable operation of the reactor can often have large, unanticipated consequences on microstructural evolution. Startup and shut down are especially important in this respect.

Data accumulated in the last few years have shown important effects of temperature history during irradiation on the behaviour of 300 series stainless steels. The effects observed have frequently been misinterpreted in the past because the experimenters assumed that irradia-

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tion had been carried out isothermally, whereas, as fact the microstructures had been greatly modified by operation for short times, at temperatures much lower than nominal.

Temperature changes in reactor core components may occur during irradiation as a result of events such as unplanned temperature excursions during power transients or scheduled relocations of fuel subassemblies. Data from ion and electron irradiations have indicated that swelling decreases when a reduction in irradiation temperature occurs if the initial temperature was below the peak swelling temperature [48]. From basic point of view very important effect of low-temperature transient exists when the transient occurs at the beginning or intermediate of the irradiation, since this could alter the microstructural evolution pathway. Nucleation of voids, loops and other radiation-induced defects typically occurs at doses < 0.1 dpa. Therefore, enhanced nucleation of voids and/or loops during a low-temperature transient at the beginning of the irradiation may significantly alter the sink strength of the evolving microstructure. This could produce either improved or degraded radiation resistance of the material compared to a steady-temperature irradiation, depending on the detailed experimental conditions.

A decrease in irradiation temperature from 520°C and 585°C at dose range 30...50 dpa causes swelling to be greater than that which occurs during constant temperature irradiation [49] (fig. 16).



Fig. 16. Enhanced swelling induced by gradual temperature reduction (ΔT) during EBR-II irradiation of 20%CW AISI 316 [49]

The effects of temperature reduction on swelling appear to be strongly dependent on the initial irradiation temperature. Reductions from initial temperatures near the peak swelling temperature are not as effective in increasing swelling as the reductions from lower initial temperature. Observed increase in swelling is can be associated with changes in precipitate morphology, but it manifest as an increase in void volume fraction. It appears that the origin of the enhanced swelling with decreasing temperature in 20 percent cold- worked AISI 316 arises from the acceleration formation of radiation induced phases (mainly γ' and G-phase) and a concur-

rent short-circuiting of the normally sluggish microchemical evolution observed at temperatures below 550°C.

Yang and Garner [50] reported that total swelling may not change an effect of a temperature change at high fluence but the void density is sensitive to decreases in temperature. The largest effects of temperature change occur during the early transient regime of swelling and are most pronounced for heats that exhibit the most resistance to swelling. The larger influence appears where never reasonably large temperature changes are made across the temperature range around 500°C.

Figure 17 shows the history of irradiation temperature of mid wall of fuel claddings irradiated in JOYO and FFTF at the center plane of reactor core. The observed difference of temperature between maximum and minimum temperature was about 30°C during irradiation period because the temperature of cladding was gradually decreasing with burning of fuel. Fuel cladding is irradiated at relatively high temperature in the initial period of irradiation. Authors [45] suggested that recovery of dislocations in cladding with the temperature decreasing progressed more than the estimated recovery of dislocation in cladding where irradiation temperature is constant during irradiation period. Seems there is some possibility that the microstructural evolution of cladding irradiated with temperature decreases is similar to the microstructural evolution of sample which has low dislocation density.



Fig. 17. Temperature history in mid wall of cladding at axial position of core center plane during irradiation in fuel assembly [45]

Experiment on BR-5 (temperature of nominal regime 430...500°C), which from the beginning changed in such regime (1 – temperature of sodium outlet was under 300°C; 2 – 300...400°C; 3 – 400...450°C) and then exploitation at 450...500°C. Void swelling of channel material (steel 18Cr9NiTi) was comparatively lower than was estimated (at 460°C and D=40dpa, S=0.1%) [51].

It was suggested that low temperature irradiation of this steel served as preliminary deformation of austenitic steels, which efficiently restrict swelling development during subsequent irradiation.

The influence of step wise temperature change on microstructural evolution and on swelling in a phosphorus modified Fe-Cr-Ni- alloy during heavy ion irradiation was investigate in work [52]. It was shown that under certain irradiation conditions the first irradiation at lower temperature strongly affects the phosphide formation during the second irradiation at higher temperature. The change in phosphide formation may result in a shortening of the incubation time for void formation.

While it appears that low temperature micro structure produced at low fluence levels will tend to dissolve at higher temperatures and higher fluences, significant micro structural alteration persisted.

CONCLUSIONS

Void swelling up to now is the main limiting factor in FBR exploitation. Analytical investigation of few environmental conditions on swelling behaviour and microstructural evolution of core and structural materials was done.

The effect of dose rate could essentially be due to the fact that, in a higher flux, a component spends less time at higher temperature in the reactor to reach a given fluence and, as consequence the achievement of a given microstructural evolution of the alloy is delayed.

- Effects of dpa rate seems operate in wide temperature interval and wide interval of dpa rate. At low temperatures this effect is sensitive to state condition of material. From the point of the evaluation of the irradiation effects on the core material and the development of the future high-performance core material, data at higher dose rates (on the order of 10⁶ dpa/sec) and higher irradiation temperatures (500... 600°C) which would be obtained in the fuel subassembly region, are more representative.
- Stress effects influenced on all components of defect structure-dislocations, voids and solid solution together with the precipitates. Stress influence is mainly pronounced in transient regime and this effect is sensitive to irradiation temperature.

Unfortunately up to now many aspects of stress influence on swelling are not understood well especially comparison between results from fuel pins and samples.

Most studies based on fuel pin cladding were inconclusive, primary because the irradiation conditions are either too complicated or poorly defined to allow a definite statement of the role of stress. Fuel pins generally operate with low initial levels of stress that increase with fluence due to accumulating fission gas pressure; thus the stress becomes large only after void incubation is over.

 Temperature history seems very important influence on swelling enhancement in fuel cladding is irradiation temperature history during lifetime, which the irradiation temperature decreased gradually with fuel burn up.

Knowledge of environmental conditions influence on microstructural behaviour and swelling can be one of the keys positions in estimating of swelling behaviour up to higher doses for achievement of high burn up of fuel. 1.F.A. Garner. Irradiation Performance of Cladding and Structural Steels in Liquid Metal Reactors *//Material Science and Technology*, A Comprehensive treatment. Vol 10A Nuclear Materials, Eds., R.W. Cahn, P. Haasen and E.J. Kramer, (VCH Weinheim, 1994)

2.A.D. Brailsford and L.K. Mansur //Journal of Nuclear Materials (JNM). 1981, v. 103&104, p. 1403.

3.Westmoreland, J.E. Sprague, J.A. Smidt et al. //Radiation effects. 1975, v. 26, p. 1–16.

4.F.A. Garner et al. //Radiation Effects and Defects in Solids. 1990, v 113, p. 229.

5.L.L. Seran and J.M. Dupouy. *Effect of time and dose rate on the swelling of 316 cladding in Phenix, Dimensional stability and mechanical behaviour in irradiated metals and alloys.* BNES, London, 1983, p. 5–8.

6.A.V. Kozlov et al. The swelling dependence of cold worked 16Crl5Ni2MoMn steel on neutron Irradiation temperature, fluence and damage rate during its use a cladding material in the BN-600 reactor. ASTM STP 1045, 2001, p. 457–468.

7.M. Kiritani //JNM. 1989, v. 169, p. 89-94.

8.T. Okita, N. Sekimura, F. Garner et al. *The primary origin of dose rate effects on microstructural evolution of austenitic alloys during neutron irradiation.* To be published in proceedings of ICFRM-10 (Baden- Baden), 2001.

9.T. Muroga, H. Watanabe, N. Yoshida //*JNM*. 1990, v. 174, p. 282–288.

10.H.R. Brager, L.D. Blackburn, D.L. Greenslade //JNM. 1984, v. 122&123, p. 332-337.

11.E. Lee, L.K. Mansur, A.F. Rowcliffe //JNM. 1984, v. 122&123, p. 299–304.

12.V.N. Voyevodin, I.M. Neklyudov, V.V. Bryk et al. Microstructural evolution and radiation stability of steels and alloys *//JNM*. 1999, v. 271&272, p. 290–295.

13.N. Azam, L. Le Naou, J Delaplace //JNM. 1973/74, v. 49, p. 197–198.

14.L.Le Naour, N. Voullon, V. Levy. *Influence of dose and dose rate on the microstructure of solution annealed 316 irradiated around 600°C.* ASTM STP 782, 1984, p. 310–324.

15.L. Boulanger, L.Le Naour and V. Vevy. Effect of dose rate on the microstructure of cold-worked 316 stainless steel, See [5], 1-4.

16.T.R. Allen., J.I. Cole, E.A. Kenik. *Radiation-Induced Segregation and void swelling in 304 Stainless Steel.* ASTM STP 1045, 2001, p. 427–442.

17.P.J. Maziasz and C.J. Mc Hargue. *Microstructural* evolution in annealed austenitic stainless steels during neutron irradiation stainless steels, International Materials Reviews. 1987, v. 32, N 4, p. 190–219.

18.V. Voyevodin. *Structural-phase changes in austenitic and ferritic stainless steels under irradiation with neutrons and charge particles*: Doctoral dissertation, Kharkov St ate University, 1995 (in Russian).

19.D. Gilbon, L.Le Naour, C. Rivera. *Effect of irradiation temperature on the precipitation in cold-worked titanium-stabilized type 316 stainless steel.* ASTM STP, 1986, p. 115–126.

20.J.M. Perks, A.P. Marwick, C.A. English. Fundamental aspects of radiation-induced segregation in Fe-Cr-

REFERENCES

вопросы атомной науки и техники. 2005. № 3.

Серия: Физика радиационных повреждений и радиационное материаловедение (86), с. 3-17.

Ni alloys. Proceedings of "Radiation-induced sensitization of stainlesssteels", Edited by P.I. Norris. London, 1987, p. 15–34.

21.J. Tenbrink, P.R. Wahi, H. Wollenberger //JNM. 1988, v. 155-157, p. 850–855.

22.J.I. Cole, T.R. Allen, S. Ukai et al. *Swelling and Microstructural Evolution in 316 Stainless Steel Hexagonal Ducts Following Long-Term Irradiation in EBR-II.* ASTM STP 1045, 2001, p. 413–426.

23.T.R. Allen, J.I. Cole, T. Yoshitake et al. *The effect of* low dose rate irradiation on the swelling of 12% coldworked 316 stainless steel, Proceedings of the Ninth International Symposium on Environmental degradation of materials in Nuclear Power Systems-water Reactors. August 1999, Newport Beach, CA, in press.

24.V. Voyevodin. The Third Review Meeting for the JNC International Fellowship Program, Tokai, 2002, p. 177–187.

25.M.H. Hassan, J.P. Blanchard, G.L. Kulcinski. Stress-enhanced swelling-Report UWFDM-901 (1992) University of Wisconsin.

26.P. Dubuisson, A. Maillard, C. Delalande et al. *The effect of phosphorus on the radiation-induced micro structure of stabilized austenitic stainless steels* ASTMSTP 1125, 1992, p. 995–1013.

27.R. Hubner, K. Erlich. Swelling and in-pile creep of neutron irradiated 15Crl5NiTi austenitic steels in the temperature range of 400 to 600°C, Influence of high dose irradiation on core structural and fuel materials in advanced reactors /Proceedings of a Technical Committee meeting IAEA Obninsk, Russia, 1997, IAEA-TECDOC-1039, p. 223–231.

28.K. Erlich //JNM. 1981, v. 100, p. 149-166.

29.K. Hubner and K. Erlich. *Influence of minor alloying el*ements and stress on microsrtuctural evolution and void swelling of austenitic stainless steels under neutron irradiation /ASTM STP, 1366, 2000, p. 778–792.

30.S. Porollo, A.N. Vorobjev, Y.V. Konobeev et al. *Irradiation Creep and Stress-Affected Swelling in Austenitic Stainless Steel 16Cr-15Ni-3Mo-Nb-B irradiated in the BN-350 Reactor.* See [29], p. 679–688.

31.J.F. Bates and E.R. Gilbert //JNM. 1976, v. 59, p. 95–102.

32.V. Shamardin, V. Golovanov, A. Povstyanko et al. *Irradiation Creep and Swelling of 16Cr15Ni3MoNb Steel and Its Modification 16Cr15Ni3MoNbB steel* //ASTM STP 1046 . 1990, v. 2, p. 753–765.

33.H.K. Sahu, P. Jung //JNM. 1985, v. 136, p. 154-158.

34.T. Lauritzen, S. Vaidyanathan, W.L. Bell et al. *Irradiation-induced swelling in AISI 316 steel: Effect of Tensile and compressive Stresses //*ASTM STP 955(VI), 1988, p. 101-113. 35.F.A. Garner, E.R. Gilbert, D.L. Porter. *Stress-Enhanced Swelling of Metals During irradiation* //ASTM STP 725, 1981, p. 680–697.

36.L.G. Walters, J.E. Flinn //JNM. 1974, v. 52, N1, p. 112–114.

37.C.C. Matthai and D.J. Bacon //JNM. 1983, v. 114, p. 22–29.

38.S. Porollo, Y.V. Konobeev, AS. Kruglov et al. *Stress-Affected Dislocation Development of Stainless Steel During Neutron Irradiation*. See [29] p. 850–859.

39.Ye. Loguntsev, V. Safonov, S. Tymentsev et al. //Fiz. *Metal. Metalloved.* 1984, v. 57, p. 802–807.

40.H.R. Brager, F. Garner, G. Guthrie //JNM. 1977, v. 66, p. 301-321.

41.N. Igata, Y. Kohno, H. Tsunakawa et al. *Influence of applied stress on swelling behaviour in type 304 stainless steel* //ASTM STP 870, 1985, p. 265–277.

42.D. Kneff, L. Greenwood, B. Oliver et al. //JNM. 1986, v. 141/143, p. 824–828.

43.R. Bullough, D. Harries, M.S. Hayns //JNM. 1980, v. 88, N2/3, p. 312–314.

44.F.A Garner and D.L. Porter. *History dependence and consequence of the microchemical evolution of AISI 316* /ASTM STP 782, 1982, p. 295-309.

45.N. Akasaka, I. Yamagata and S. Ukai. *Effect of Irradiation Environment of Fast Reactor's Fuel Elements on void swelling in P,Ti - Modified 316 Stainless Steel* /ASTM STP 1045, 2001, p. 443–456.

46.H. Brager, F. Garner, E. Gilbert et al. *Stress-Affected Microstrucurural development and the Creep-Swelling Interrelationship In Proceedings: Radiation Effects in Breeder Reactor Structural Materials*. Arisona, 1977, p. 727–756.

47.B.J. Makenas, J.F. Bates, and J. W.Jost. *The swelling behaviour of 20% CW 316 Stainless Steel Cladding Irradiated with and without Adjacent Fuel* /ASTM STP 782, 1982, p. 17–30.

48.F. Garner, N. Sekimura, M. Grossbeck et al. //JNM. 1993, v. 205, p. 206–218.

49.J.F. Bates and D.S. Gelles //JNM. 1978, v. 71, p. 365.

50.W.J. Yang and F.A. Garner. *Relationship between phase development and swelling of AISI 316 during temperature changes* /ASTM STP, 782, 1982, p. 186–206.

51.S. Porollo, V. Sherback, N. Aristarchov et al. Influence of exploitation regime of BOR-5 reactor on void swelling in 18Cr9NiTi steel *//Atomic Energy*(in Russian), 1977, v. 43, p. 206–207.

52.D. Hamaguchi, H. Watanabe, T. Muroga et al. //*JNM*. 2000, v. 283-287, p. 319–323.

ВЛИЯНИЕ ВНЕШНИХ ФАКТОРОВ НА МИКРОСТРУКТУРНУЮ ЭВОЛЮЦИЮ И РАСПУХАНИЕ В ОБЛУЧЕННЫХ АУСТЕНИТНЫХ НЕРЖАВЕЮЩИХ СТАЛЯХ

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Исследованы и проанализированы внешние условия, влияющие на поведение распухания облученных аустенитных нержавеющих сталей. Изучено влияния скорости создания смещений, внешних напряжений и температурной истории на микроструктурную эволюцию и распухание в этих сталях.

ВПЛИВ ЗОВНІШНІХ ФАКТОРІВ НА МІКРОСТРУКТУРНУ ЕВОЛЮЦІЮ ТА РОЗПУХАННЯ В ОПРОМІНЕНИХ АУСТЕНІТНИХ НЕРЖАВІЮЧИХ СТАЛЯХ

В.М. Воєводін, Ф. Гарнер, М.П. Лазарєв, О.С. Кальченко, Н.А. Удалова

Досліджені і проаналізовані зовнішні умови, що впливають на поведінку розпухання опромінених аустенітних нержавіючих сталей. Вивчено вплив швидкості створення зсувів, зовнішніх напруг і температурної історії на мікроструктурну еволюцію і розпухання в цих сталях.