## **SECTION II**

## **REVIEW ARTICLES**

#### **REVIEW ARTICLE**

# IRRADIATION CREEP AND INTERRELATION WITH SWELLING IN AUSTENITIC STAINLESS STEELS

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The available experimental data on irradiation-induced creep in austenitic stainless steels are summarized and the existing theories reviewed. Attention is paid to the influence of material composition and pretreatments on irradiation creep. In particular the stress, flux, fluence and temperature dependencies are reported and possible correlations of irradiation creep with the microstructural evolution, the swelling behaviour and the precipitation kinetics of the materials are outlined. The consequences of stress effects connected with swelling for the irradiation-creep behaviour, especially the stress-dependence, are discussed.

#### 0. Introduction

Austenitic stainless steels have been chosen worldwide as standard cladding and subassembly wrapper materials for prototype and commercial fast breeder reactors [1–8]. The initial choice of materials was made on the basis of "classical" properties such as creep properties, corrosion behaviour in liquid sodium, and nuclear data which influence the neutron absorption and the breeding ratio.

In the meantime the phenomena of void swelling and irradiation creep were observed in non-fissile metals [9,10], hence the consequences of dimensional instabilities for the core constructions induced by irradiation creep and swelling were analysed [e.g. 11,12]. These studies generally showed that irradiation creep and swelling of the materials may limit the achievable burnup. This maximum burn-up is very much dependent upon the exact circumstances and the correlations used for these irradiation effects and the method used to extrapolate them to high neutron fluences.

The general opinion of the fast breeder community is, however, that the material requirements for the prototype fast reactors (Phénix, PFR, BN 350, SNR 300, MONJU and the test facility FFTF) can be fulfilled by the reference materials. Especially the very successful operation of Phénix with about 10000 fuel pins which have accumulated a neutron exposure of 65 dpa-NRT or more [13], are a strong support for this opinion.

The question whether or not commercial fast breeders with much higher neutron damage (85–90 dpa) can be constructed by using reference austenitic stainless steels as core structural materials is still open. This is, however, not a principal restriction of the fast breeder technology since a further optimization of reference materials and their partial replacement by alternative materials (e.g. ferritic/martensitic steels or Ni-base alloys) are the aims of intense research programmes all over the world.

# 1. Reference cladding and sub-assembly wrapper materials

In table 1 a list of reference cladding and subassembly wrapper materials is given for the different fast breeder projects. Typical analyses of these materials (in wt%) are collected in table 2. With the exception of Nimonic PE 16, which is already a Ni-base alloy, all the other materials are austenitic stainless steels with good sodium compatibility and high creep strength at the operational temperature of 400–650°C. As can be seen, some of these steels are unstabilized (AISI 316, M316); types 1.4981, 0Cr16Ni15Mo3B and FV 548 are niobium-stabilized and types 1.4970 (12R72HV), AISI 321 and 1Cr18Ni10Ti are titanium-stabilized. In addition there exist titanium-modified versions of AISI 316.

A guideline for the austenite stability or the possibil-

Country	Reactor	Cladding material	Sub-assembly wrapper
DEBENE a)	SNR 300	1.4970	1.4981
France	Phénix	AISI 316	316 L
	Superphénix	316 Ti	316 Tı
JAPAN	Monjou	316	316
UK	PFR	M 316 (FV 548)	Nimonic PE 16 (AISI 321)
	CDFR	316	PE 16
USA	FFTF	AISI 316	AISI 316
USSR	BN 350	0 Cr 16 Ni 15 Mo 3 B	1 Cr 18 Ni 10 Ti
	BN 600		

Table 1 Core reference fuel element structural materials for prototypic and commercial fast breeders

ity of  $\delta$ -ferrite formation, respectively martensitic transformation, for the different alloys is the Schaeffler diagram [16] which has been initially used to predict the effect of the composition on weld-metal constitution. We use it in an extended form to characterize the stability of reference materials; it incorporates the N content [17]. According to fig. 1, type 1.4970 is the most stable alloy with respect to  $\delta$ -ferrite formation, respectively martensite transformation.

A further important criterion for the application of these alloys is their precipitation behaviour. Two aspects have especially to be taken into account: the direct influence of precipitates and their interfaces on the formation and the changes in chemical composition which accompany their formation. A large variety of intermetallic phases and carbide precipitates appears in different austenitic stainless steels. Among those intermetallic phases which are possible in iron-base alloys,

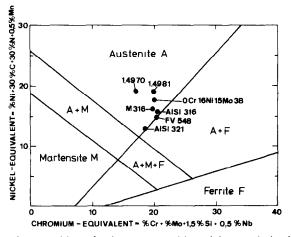


Fig. 1. Position of reference austenitic stainless steels in the Schaeffler diagram [16,17].

(fig. 2) [18,19], the  $\sigma$ ,  $\chi$  and Laves phases are of practical importance. Especially the formation of the  $\sigma$  phase was correlated very early on [20] with a reduced swelling resistance of the austenitic stainless steels. According to their position in a duplex  $\gamma + \sigma$ -field, these alloys are susceptible to  $\sigma$  phase formation, at least at 650°C and below. Again steels of type 1.4970 or 0Cr16Ni15Mo3B are less sensitive than, for instance, AISI 316. In practice,  $\sigma$ -phase formation plays no role in the Ti-stabilized material 1.4970 during thermal annealing at the operation temperature of 400–650°C.

Finally, all austenitic stainless steels are susceptible to the formation of carbide precipitates in form of  $M_{23}C_6$ ,  $M_6C$ ,  $M_7C_3$  (M=Cr, Mo, Fe or Ni) which form at grain boundaries as well as intragranularly. In case of stabilization by titanium or niobium additions carbides of type MC (M=Ti, Nb) appear mainly intragranularly in the matrix. The stabilized steels mentioned here are in general fully stabilized, e.g. carbon can be bound by the elements Nb and Ti.

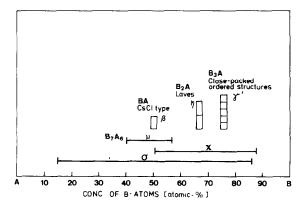


Fig. 2. Intermetallic phases which may form in austenitic stainless steels [18,19]; (A=Ti, V, Cr, Nb, Mo), (B=Fe, N1, Mn, Co).

a) DEBENE: Abbreviation for Deutschland, Belgium, The Netherlands.

Analyses (wt%) of more common reference clad and wrapper structural materials for prototypic and commercial fast breeders

1 1

Denomination	C	Si	Mn	Ç	ï	Mo	Ti	N <sub>p</sub>	z	В	Ref.
AISI 316 (1.4919)	0.08	<0.75	1.5-2.0	1.5–2.0 16 –18	12 -14	2 -2.5		l	≤0.01	€0.001	[3]
M 316	0.03-	9.0≥	1.5-2	15.4–17.5 13 –14	13 -14	2 -2.75	0.05 max.	0.05 max.	≤0.02	0:001	[5]
1.4981	0.04-	0.3-0.6	<1.5	15.5-17.5	15.5–17.5 15.5–17.5	1.6–2.0	1	0.4-1.2	0.01	≤0.001	4
0 Cr 16 Ni 15 Mo 3 B	0.05-	9.0	≥0.8	15 -17	14 - 16	2.5-3	ı	6.6-0.9	1	0.005	[14]
1.4970	0.08- 0.12	0.3-0.75	≤2.0	14.5–15.5	14.5–15.5 14.5–15.5	1 -1.4	0.33 - 0.55	I	≤0.015	0.003- 0.008	<u>4</u>
Nimonic PE 16	0.05-	≤0.3	<0.29	15 –18	42 –45	2.5–4	1.1 –	A1 1.1–1.5	0.011	0.001	[5]

### 2. Irradiation creep mechanisms

Irradiation creep is caused when external non-hydrostatic stresses are applied during irradiation. There are a number of theoretical approaches proposed in the literature to explain this phenomenon. They will briefly be summarized especially with respect to their possible intercorrelation between irradiation creep and swelling. For a more detailed discussion the reader is referred to recent conference reports [1,2,21].

The mechanisms of stress-induced loop nucleation or alignment (SIPN) and stress-induced preferrential absorption (SIPA) have as a common feature that the formation or growth of interstitial loops or the absorption of defects at existing dislocations depends upon their orientation with respect to the external stress and therefore cause a macroscopic creep strain. Glide processes are hereby neglected.

Those models which include dislocation glide (climbenabled glide) as a further strain contribution are of especial interest in cases where the linear stress-dependence of creep deformation is no longer valid or where the creep rate increases at the onset of swelling.

# 2.1. Stress-induced loop nucleation or loop alignment (SIPN)

Stress-induced loop nucleation or loop alignment of interstitial clusters was initially proposed by Hesketh [22] as an important mechanism for irradiation creep. The corresponding model is based upon the assumption that irradiation-induced interstitial loops are preferentially nucleated on crystallographic planes oriented nearly perpendicular to the applied stress. Garner et al. [23] explored this possibility of stress-induced loop nucleation by adapting the void nucleation theory of Katz, Wiedersich and Russell to this problem. Since there exists obviously no nucleation barrier for clusters larger than di-interstitials one would therefore not expect an effect of stress on their nucleation. Early experimental observations of different loop densities on different {111} planes in stressed type AISI 316SS [24] could better be explained by a stress-induced reorientation of small interstitial clusters [25]. Such a possibility of stress effects on the orientation of nuclei has been indeed found in aluminium by Robrock et al. [26].

The idea of Hesketh's stress-induced loop nucleation was developed further by Lewthwaite [27], Herschbach and Schneider [28] and later Brailsford and Bullough [29] in order to predict the irradiation creep rate,  $\dot{\epsilon}$ , Brailsford and Bullough deduced the following equation:

$$\dot{\varepsilon} = \frac{2}{3} \frac{\sigma b^3 n}{3kT} \frac{\rho_{\rm D}^{\rm L}}{\rho_{\rm D}} \dot{S} \tag{1}$$

with  $\sigma$  as the applied stress, b a measure of interatomic distance, n the number of interstitial atoms forming a critical nucleus,  $\rho_D$  the total dislocation density,  $\rho_D^L$  the dislocation density of interstitial loops and  $\dot{S}$  the swelling rate.

The application of this model is, according to Bullough [30], restricted to materials with low dislocation densities, e.g., solution-annealed materials at low neutron fluences.

### 2.2. Stress-induced preferential absorption (SIPA)

Stress-induced preferential absorption of interstitials at dislocations has been proposed by Heald and Speight [31] and Wolfer and Ashkin [32]. The origin of the SIPA mechanism is the elastic interaction of a point defect with an edge dislocation. The magnitude of this interaction is quadratic in the total strain-field. If an external stress is applied in addition to the strain field of the dislocation, this superimposed strain field leads to interaction energies which depend on the orientation of the edge dislocation.

The treatment of Bullough and Willis [33], and Heald and Speight [34] concentrated on the special case where a uniaxial stress is applied parallel, respectively perpendicular to the Burgers vector of an edge dislocation. Bullough and Hayns [35] calculated the irradiation creep strain as:

$$\varepsilon \sim 0.5 Kt(\sigma/G),\tag{2}$$

where K = displacement rate, t = time, and G = shear modulus. According to this result, the creep rate is essentially independent of the irradiation temperature, the dislocation density and other material parameters. A temperature dependence through the thermal emission of vacancies from vacancy loops generated in displacement cascades has been proposed by Bullough and Hayns [36].

Wolfer [24] applied the perturbation treatment to this problem and derived the following equation for the SIPA-creep rate.

$$\varepsilon_{\text{SIPA}} = \sigma \left[ N^{\text{L}} \xi^{\text{L}} \Delta Z^{\text{L}} + N^{\text{D}} \xi^{\text{D}} \Delta Z^{\text{D}} \right] F, \tag{3}$$

where  $N^{L}$  and  $N^{D}$  are the concentrations of loops (L) and dislocations (D),  $\xi^{L}$  and  $\xi^{D}$  corresponding proportionality factors,  $\Delta Z^{L}$  and  $\Delta Z^{D}$  the stress-induced net bias for loops and dislocations, F a complex parameter which contains the defect rate K.

2.3. Irradiation creep by climb-enabled glide of disloca-

Mansur and Reiley [37] very recently summarized the models which have been proposed to explain irradiation creep by climb-enabled glide of dislocations. Two distinct lines in the development of theories can be seen. Those models which connect the climb of dislocations with the differences in point defect capture efficiencies of different types of sinks (dislocations, voids, grain boundaries, free surfaces, precipitate—matrix interfaces) are the historical ones. In most cases the net interstitial flux to dislocations has been connected with the swelling [38,39].

Most of the authors have employed a model of deformation by dislocation glide after climb which had been developed by Ansell and Weertman [40] in order to explain thermal creep in disperion-hardened aluminium alloys. For shear stresses where Frank—Read dislocation sources are operating, the creep rate  $\dot{\varepsilon}$  is proportional to the swelling rate  $\dot{S}$  and shows a quadratic stress dependence.  $\lambda$  is the mean distance between the hardening obstacles:

$$\dot{\varepsilon} \sim (\dot{S}\lambda/G^2)\sigma^2. \tag{4}$$

For lower stresses Wolfer et al. [41] have developed a model in which dislocation loops act as obstacles. The stress dependence of the creep rate is then parabolic at low stresses and linear at higher stresses.

Gittus [39] proposed a dislocation climb and glide model, termed I-creep, which is analogous to the Cottrell creep mechanism in  $\alpha$ -uranium. Edge dislocations climb by absorption of excess interstitials into regions where they can bow out under the influence of the externally applied stress.

The amount of bowing of the dislocations is restricted to a configuration where the line tension force of the dislocation equals the applied stress. This fractional bowing was estimated by Mott [42] and Friedel [43] to be  $\sigma/E$ . A continuation of climb and glide processes then leads to a creep rate formulated as

$$\dot{\varepsilon} \sim \dot{S}\sigma/Y,$$
 (5)

where Y =yield stress.

All these models of irradiation creep by climbenabled glide of dislocations need the net flux of interstitials as a consequence of vacancy loss to other sinks (e.g. voids etc.).

Mansur [44] finally introduced the idea of the stressinduced preferential absorption (PA) and emission (PE) of the radiation-generated defects into the climb-glide

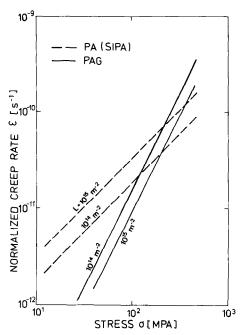


Fig. 3. Stress dependence and domains for SIPA and PAG creep [37].

model. It is denominated as PAG-creep. The complete model includes both the unequal partitioning of point defects among the dislocations themselves and the net interstitial absorption at dislocations corresponding to the net vacancy flow to other sinks.

For the simple case of neglecting all other sinks other than the dislocations and neglecting further the emission terms (PE), the creep rate by preferential absorption and glide (PAG) is given as:

$$\dot{\varepsilon}_{\rm PAG} = \frac{4\Omega}{9h} (\pi L)^{1/2} \Delta Z^{\rm D} D_{\rm J} C_{\rm J} \frac{\sigma^2}{F}$$
 (6)

with L as the dislocation line density,  $D_j$  and  $C_j$  the diffusion coefficient and concentration of interstitials respectively, and  $\Omega$  atomic volume.

The SIPA and PAG-creep models differ mainly with respect to their stress dependence and a weaker influence of the dislocation density in the latter theory. Accordingly, SIPA creep dominates at high dislocation densities and low stresses (fig. 3).

### 3. Experimental results on irradiation creep

In the temperature region 400-500°C, where bulging of the wrapper tubes occurs owing to differential sodium

hydraulic stresses at the bottom of the wrappers, the irradiation creep contributes a dominant part to the deformation of a subassembly. This is the reason why most of the experiments have been performed in this temperature range. Subsections 3.1 and 3.2 will deal with this range, whereas 3.2 will concentrate on the effect of temperature on irradiation creep.

In order to compare the different experiments the following normalizations are used: the neutron fluence is always given in dpa (displacements per atom, according to the NRT model). Hence dpa(F), the French scaling of dpa and dpa(N/2), the Half-Nelson calculation, are multiplied by a conversion factor of 0.77, respectively 0.8 [45,46].

Since different test-methods were used to measure the irradiation creep (e.g. Uniaxial tests, UT; Pressurized Tube Tests, PTT; Helical Spring Tests, HST), the data are generally converted into the effective strain  $\bar{\epsilon}$  and stress  $\bar{\sigma}$  values by the Soderberg formalism [47]:

$$\bar{\varepsilon}/\bar{\sigma} = \varepsilon/\sigma = \gamma/3\tau = 4\varepsilon_{\rm H}/3\sigma_{\rm H},\tag{7}$$

where  $\varepsilon/\sigma$  are tensile,  $\varepsilon_{\rm H}$ ,  $\sigma_{\rm H}$  are hoop, and  $\gamma$ ,  $\tau$  are surface shear strains, respectively stresses. The stress-normalized strains are used in those cases where the experimenters explicitly reported a linear relation between applied stress and strain.

The data are also corrected for the swelling of stress-free samples, e.g. for PTT,  $\varepsilon_{\rm H} = \varepsilon_{\rm TOT} - \varepsilon_{\rm S}$ , where  $\varepsilon_{\rm TOT}$  is the total diametral change under stress and  $\varepsilon_{\rm S}$  the diametral change due to the swelling of a corresponding stress-free sample.

Correction for an additional stress-induced swelling could generally not be introduced, since there exist only few experimental data. In case of HST one should theoretically have the possibility to eliminate this additional effect since the hydrostatic stress component is zero in this stress system and hence swelling should not be affected.

#### 3.1. Irradiation creep in AISI 316

In fig. 4 data of neutron experiments with AISI 316 in a 20% cold-worked condition are compiled [48-52]. If one takes into account that different types of experiments and different heats, even a slightly modified alloy (M316), have been used, the agreement between the different results is surprisingly good. Since the temperature vary between 280 and 500°C one can conclude the irradiation creep is only weakly temperature-dependent.

With few exceptions [48,50] irradiation creep rates increase with the neutron fluence. Hence a "creep coef-

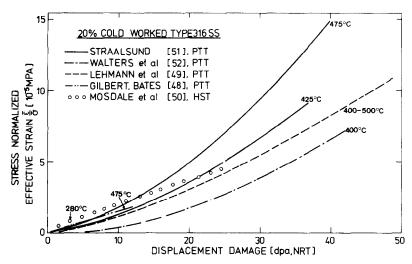


Fig. 4. Dose dependence of the stress-normalized creep deformation for Type AISI 316 CW.

ficient" 
$$\overline{K}$$
, defined by

$$\varepsilon/\sigma = \overline{K}\phi t, \tag{8}$$

is dependent upon the neutron flux, the fluence and the temperature:  $\overline{K} = \overline{K}(\phi, t, T)$ . We will nevertheless use such a creep coefficient, which normalizes the creep strain with respect to the stress and the neutron fluence, given in dpa, in order to compare different materials at low neutron fluence levels.

For low dose irradiations the published data indicate (fig. 5) very similar  $\overline{K}$  values for type AISI 316, 20% CW, in the temperature range 280–475°C. They are in the order of  $1 \times 10^{-6}$  MPa<sup>-1</sup> dpa<sup>-1</sup> with nonsys-

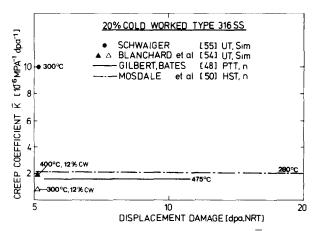


Fig. 5. Dose dependence of the creep coefficients  $\overline{K}$  for type AISI 316 CW in low-dose irradiations.

tematic scatters of less than  $\pm 1 \times 10^{-6}$  MPa<sup>-1</sup> dpa<sup>-1</sup>. One exception exists [59] with a value in the range of  $10^{-5}$  [MPa<sup>-1</sup> dpa<sup>-1</sup>], however, this might well be due to the special material employed, namely, a "pure" type 316SS.

In most cases the creep strain per unit dose appears to be independent of the dose rate, which implies a linear correlation between damage rate and creep rate. This has been confirmed by simulation experiments with protons [55–57] for nickel, AISI 304 and AISI 316 at about 300°C. Walters et al. [52] and Dupouy et al. [58] come to the same conclusion for the austenitic steels AISI 304 and 316 after neutron irradiation. However, Lewthwaite and Mosdale [59] report that in their investigations in DFR the strain per unit damage dose decreases as the dose rate increases, and they exclude that this is a temperature dependent effect induced by trapping. Similar effects have been observed in PE 16 [60] and in ferritic steels [61].

In general a linear stress-dependence of irradiation creep has been reported in the publications mentioned above.

However, in a few cases deviations from this linear correlation were observed [49,51,53] especially at high stress levels. The stress exponents n increase to values which might be expected for a thermal creep mechanism.

This relatively uniform creep behaviour of AISI 316, 20% CW, under neutron irradiation has recently been clouded since Gilbert and Lovell [62] found a remarkable difference in the creep deformation of "N-lot" and "NICE"-lot, two different batches of type 316 stainless

steel, the latter showing much higher creep strains per dose unit. Since the NICE-lot also exhibits higher thermal creep rates, Gilbert and Lovell interpret this as a direct interrelation between the thermal and the irradiation creep. Recent investigations of type 316 (chemically modified with several solutes) revealed another very interesting aspect. According to Bates et al. [63] those solutes which reduce swelling in AISI 316 also reduce the creep strain under irradiation (fig. 6). Several mechanisms have been discussed to explain this effect, such as "fast diffusing species" [64], trapping effects [65], or the direct interaction of the solute atoms with dislocations which causes a change in their bias factor.

Thermochemical treatments of AISI 316 change the irradiation creep, as has been shown by Walters et al. [52] (fig. 7). At 400°C the measured creep deformations are higher for the cold-worked material than for the solution-annealed steel. A thermal preageing at 704°C/216 h which is accompanied by the formation of coarse carbide precipitates at grain boundaries and slip planes, accelerates the irradiation creep. At 550°C both heat-treatments show the same deformation mode at moderate stresses.

Other data in the literature do not agree with the above observed enhancement of irradiation creep by the introduction of cold working. Lehmann et al. [49] measure higher creep rates for the solution-annealed type 316 when compared to the cold-worked steel. Mosdale et al. [50] present data on M 316 where for 280°C the steady state creep constants scatter by a factor of two for different batches and do not allow a definite conclu-

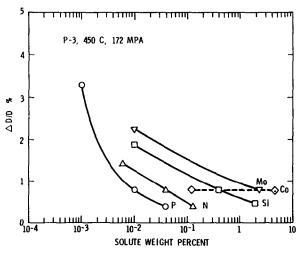


Fig. 6. Effect of minor chemical additions on irradiation creep in type AISI 316 SS [63].

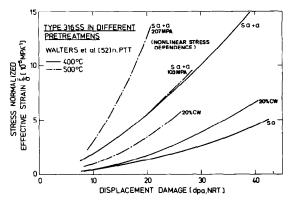


Fig. 7. Influence of thermochemical pretreatment on irradiation creep behaviour in type AISI 316 SS.

sion as to whether or not cold working enhances or reduces irradiation creep.

There is, however, agreement of the experimental observations with respect to thermal preageing. Dupouy et al. [66] reported very early that preageing before irradiation or an accelerated precipitation of  $M_{23}C_6$  carbides during irradiation enhances both creep and swelling. Hofman et al. [67] conclude that the most important parameter influencing swelling and creep of type 316 SS is the denudation of the matrix from carbon via the formation of the  $M_{23}C_6$  phase. They show, by discussing the detailed kinetics of carbide precipitates, how the temperature dependence of irradiation creep will be changed owing to the temperature variation of the amount of precipitates.

#### 3.2. Irradiation creep in other austenitic stainless steels

Type 1.4981 is, according to table 1, a niobium-stabilized 16 Cr-16 Ni steel and has been chosen as wrapper material for SNR 300. Two sets of experiments are the basis for our knowledge about irradiation creep in this material: (1) uniaxial creep tests in the BR-2 in Mol (Mol-5 B) [68,69] and (2) pressurized tube experiments in Rapsodie (RIPCEX I) [70-72]. The results of both experiments are given in the same manner as for AISI 316 in fig. 8. Again it is evident that at low neutron doses a quasi-linear dependence of creep deformation as function of fluence can be observed. In addition the BR-2 experiments indicate that there is, if any at all, only a small temperature dependence of IPC in the range between 350 and 550°C.

After the initial phase with an instantaneous creep rate in the range of  $10^{-6}$  MPa<sup>-1</sup> dpa<sup>-1</sup>, one observes a transient creep state with quickly increasing creep rates

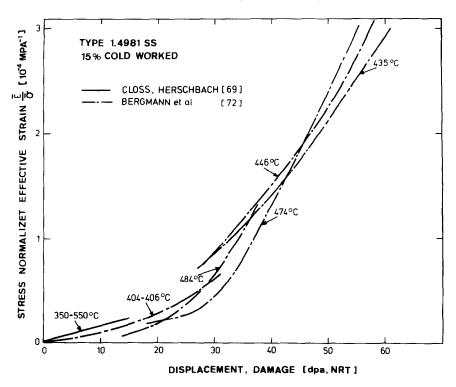


Fig. 8. Dose dependence of the stress normalized creep deformation for type 1.4981 SS (15% CW).

and finally a quasilinear steady-state regime in which the creep rate achieves values of an order of  $10^{-5}$  MPa<sup>-1</sup> dpa<sup>-1</sup>. This value is slightly dependent upon the irradiation temperature.

The analysis of stress-dependence of irradiation creep in this material can be describes as follows:

For the Mol-5B experiments the stress-dependence of the creep rate is linear in the region between 350 and  $550^{\circ}$ C and for damage levels  $\leq 15$  dpa. For the experiment with pressurized tubes (RIPCEX I) one can determine a temperature damage domain at which for a stress of  $\leq 150$  MPa a linear stress-strain correlation is obtained (fig. 9).

For stresses above 150 MPa deviations from a linear stress-dependence are observed, especially when higher neutron fluences have been accumulated (fig. 9). This apparent deviation can at the best be explained by the additional effect of a stress-driven swelling on the dimensional changes [73]. At 430 and 480°C such a limitation of the area where a linear stress-strain correlation is valid has not been observed. This might possibly be due to the low neutron-fluences achieved.

A second important feature of this material is the pronounced change of the creep rate at the onset of swelling. This implies an interrelation of the two phenomena [73] and will be discussed later. There might be a different possibility to interpret this finding. In earlier experiments [74] as well as in the RIPCEX-samples an intense formation of carbides during neutron irradiation has been observed. Unfortunately, owing to the lack of intermediate destructive post-irradiation examinations, it is not possible to investigate, whether the intense carbide formation coincides with the beginning of swelling and the enhanced irradiation creep or not. However, the suggestion that an intense formation of carbide precipitates is accompanied by an enhanced creep and swelling rate parallels the reported observations of Dupouy et al. [66] for type AISI 316 and supports their hypothesis that the denudation of the matrix of carbon is the underlying mechanism.

There are data available from other austenitic stainless steels such as FV 548, AISI 321, AISI 304 and AISI 316 [49,50,59,75]. In addition, results on the nickel-based alloys PE 16 and Inconel 706 [53,60] and on ferritic and ferritic/martensitic steels [53,76,77] have been published recently.

A comparison on the basis of these data is possible for low neutron fluences ( $\leq 20$  dpa) and in stress-regions

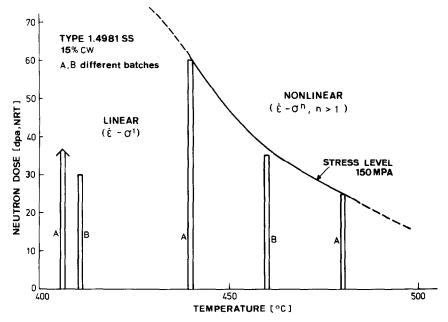


Fig. 9. Temperature/neutron-fluence domain for linear in-pile creep behaviour in type 1.4981 SS.

where—neglecting the initial transient creep phase—a linear stress and fluence dependence has been observed. In case of precipitationhardened alloys only "simple" pretreatments (e.g. solution-annealed or cold-worked materials) were selected for this comparison in order to avoid the additional effect of preformed precipitates. The creep coefficients  $\overline{K}$  have been determined in the usual manner and are plotted in fig. 10.

Paxton et al. [53] concluded from their data-base that for  $545^{\circ}$ C a clear range of order exists, in which the solid-solution strengthened austenitic and ferritic steels show the highest creep rates follwed by  $\gamma'$  hardened alloys. The  $\gamma''$  precipitation-hardened alloys exhibit the lowest values. One has, however, to add that the advantages of the Ni alloys at that temperature might be due to the effect that the solution-strengthened austenitic

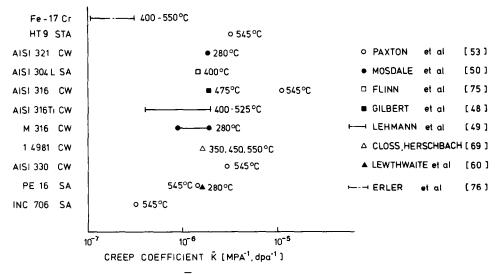


Fig. 10. Review of creep coefficients  $\overline{K}$  for a number of austenitic stainless steels.

stainless steels and the ferritic/martensitic steel HT 9 already show thermal creep at 545°C. The nonlinear stress dependence which has been found for these materials is indicative for such an assumption.

In addition, the data on Fe-17 Cr [76], a pure ferritic alloy and 316 Ti CW [49] behave similar or even better than Inconel 706 SA. Therefore we would conclude that further experimental data are necessary to evaluate the materials with the lowest irradiation creep properties.

### 3.3. Temperature dependence of irradiation creep

The data of irradiation-creep discussed so far, reveal a weak temperature dependence between 400 and 550°C for most of the austenitic steels, at least for fluence regimes where swelling is still neglibigible. The observed creep coefficient  $\overline{K}$  is in the order of  $1 \times 10^{-6}$  [MPA<sup>-1</sup> dpa<sup>-1</sup>], a value which is to be expected if the SIPA model is applied.

In experiments carried out at lower temperatures, e.g. in Mosdale's helical spring tests in DFR [59] or in simulation tests [54–56], similar creep strains per unit damage dose were obtained. Dependent upon the material, the creep strains differ by a factor of 1.5 to 5 between 280 and 500°C.

According to eq. (2) above, derived by Bullough and Hayns [35], SIPA creep should only be weakly dependent upon the irradiation temperature for T < 450°C. However, the extrapolation to temperatures of 300°C needs the introduction of the recombination of point defects as an important parameter. In this case, a steeper temperature dependence than actually observed is predicted.

The temperature dependence of irradiation creep in type 316, cold-worked, shows a fine structure, as can be seen from the data of Gilbert and Lovell [62] and Dupouy et al. [49] for type AISI 316 20% CW. In the temperature region where thermal creep is negligible the increase of irradiation creep with temperature is very moderate (see fig. 11); the creep rate goes through a maximum and shows a dip near the boundary of the domain where thermal creep becomes important. This behaviour can essentially be understood by the development of the loop and dislocation densities during irradiation [78]. Wolfer showed that the appearance of such a dip can well be attributed to a reduced dislocation density which indicates the beginning of recovery. Bullough and Hayns [36] also investigated the temperature dependence of irradiation creep. They explained the intermediate maximum in the creep deformation curve by an extensive emission of vacancies from

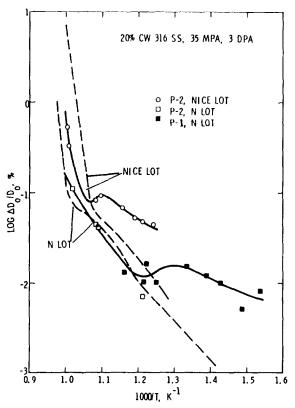


Fig. 11. Temperature dependence of irradiation creep in type AISI 316 CW [62].

vacancy loops; the latter are formed from the displacement cascades during neutron irradiation.

Only few data are available on the high-temperature deformation mode under irradiation. In this temperature region (T > 550°C) thermal creep should dominate, though changes in the microstructure can change the creep rates remarkably. From fig. 11 one can deduce that the transition temperature which separates the two creep domains in AISI 316 is at about 550°C [56]. A similar value is reported for AISI 321 [62]. However, there is ample evidence that this transition point might depend upon slight chemical variations (N-Lot of type 316 compared to NICE lot) and upon the neutron fluences achieved [48,62].

There are some discrepancies concerning the absolute values of in-reactor creep deformations when compared to corresponding thermal creep rates. Gilbert and Lovell [62] find at 700°C a reduced in-reactor creep deformation rate for type AISI 316 SS compared with data measured with pre- or post-irradiation creep tests. They explain this by assuming a dynamic hardening

effect which is caused by irradiation-induced short-lived obstacles.

Closs and Herschbach [68,69] report a more complex behaviour for type 1.4981 SS (15% CW) at 700°C. Their data originate from uniaxial tests with continuous length measurements. They, therefore, allow a more precise continuous determination of the in-reactor-creep deformation. According to fig. 12, the creep curves for unirradiated samples and in-reactor deformed samples are identical at a high stress level (120 MPa). The irradiation effect is a classical one, a reduction in total strain. Owing to low neutron exposure and high irradiation temperature matrix hardening is negligible. For a low stress level (60 MPa), however, the creep rate is enhanced for the in-reactor deformation. The authors explained this enhancement by an irradiation-induced recovery of the cold-worked structure or, alternatively, by an irradiation-enhanced coarsening of precipitates rather than by enhanced in-reactor creep. In other words, the different behaviour of the material at high or low stress levels does not indicate different creep mechanisms, but is due

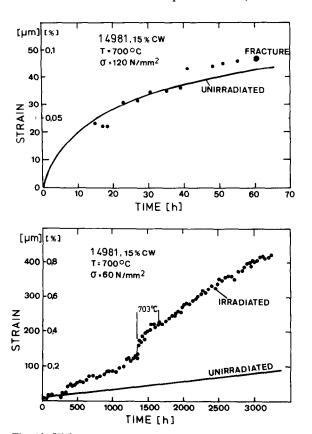


Fig. 12. High temperature deformation for type 1.4981 SS at 720°C [69].

to the different exposure times at which eventually recovery or precipitation under irradiation changes the initial structure.

Finally, Wassilew et al. [79] have reevaluated these data and extended their analysis to pressurised tube tests (Mol 2) for different austenitic stainless steels irradiated in the BR-2 reactor at 710°C. According to fig. 13 they observe in type 1.4970 for stresses < 120MPa an enhanced creep deformation under irradiation when compared with thermal, or post-irradiation creep rates. In addition they found for stresses < 120 MPa that the average creep rates under irradiation depend linearly upon the applied stress. For higher stresses, the stress-dependence of the deformation rate is identical to that found for the unirradiated material with a stress exponent n much larger than one. They could also show that for the low-stress region the creep rates depend upon the square root of the defect production rates. Especially this observation led the authors to the conclusion that there also exists irradiation creep at T > 0.5 $T_{\rm M}$ ,  $T_{\rm M}$  being the melting point for the austenitic stainless steels. The stress level which separates the two domains is dependent upon the material (120 MPa for type 1.4970 and 80 MPa for type 1.4981 SS at 710°C) and on the temperature.

This type of irradiation-enhanced creep deformation

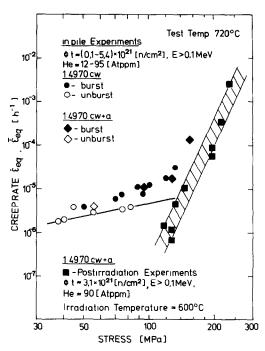


Fig. 13. In-reactor creep deformation for type 1.4970 SS at 720°C [79].

deviates, however, in one essential point from that observed at lower temperatures. The samples come to rupture at quite low ductility values, which means that an additional failure mechanism exists. A further difficulty in explaining this finding is that a linear stress dependence of creep deformation at this stress-temperature region is no longer an unequivocal proof for an in-pile-creep mechanism, as it is at lower temperature. It also could be attributed to Coble creep or Nabarro creep.

Bullough and Harries [80] have recently argued that such a type of enhanced creep might be caused by the growth of gas bubbles on the grain boundaries perpendicular to the stress axis where the residual interstitials form suitably oriented interstitial dislocation loops which extend the grain boundaries. They call this a stress-induced gas-driven irradiation creep mechanism. Further theoretical and experimental investigations have to be done to explain and clearify this very interesting field of high-temperature deformation under irradiation.

# 4. The influence of stress on the microstructural development

The microstructure, expecially the dislocation structure and its development plays an important role in the interpretation of irradiation-induced creep. As already explained in section 2, the SIPA-model is based on the assumption that by the stress-induced preferential absorption of interstitials an anisotropic, stress-dependent growth of interstitial loops or climb of dislocations occurs. One would therefore expect anisotropic distributions of these species under the combined influence of stress and irradiation.

Several investigators (Brager et al. [23,81,88]) very carefully studied the influence of stress on the loop number and loop size in AISI 316 SS. They showed that the loop number density increases with stress for lattice planes normal to the applied stress irrespective of the pretreatment of the material (fig. 14).

The enhancement of the loop number density in lattice planes normal to the applied stress is over-compensated by a decrease in number for loop orientations nearly parallel to stress. This is an important evidence for the SIPA mechanism. The loop size distributions, however, are not changed by an applied stress. This finding does not imply that the growth rates of interstitial loops would be unaffected by external stresses. It may be caused by the interaction of loops or by the reaction of loops with an existing dislocation network when the concentration and/or size of the

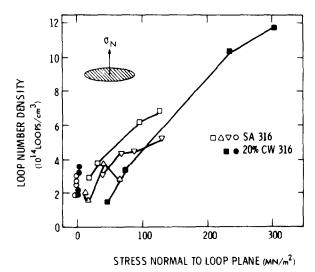


Fig. 14. The stress dependence of loop formation in type AISI 316 [81].

loops arrives at a critical value. This, indeed, happens at rather low neutron fluences, so that the loop size distribution in highly irradiated materials has only a limited value for the discussion of stress effects on the loop growth.

The development of a network dislocation structure under irradiation has been followed only in very few experiments. The results indicate in fig. 15 [82] that, irrespective of the mechanical pretreatment, the total dislocation density saturates after a relatively low neu-

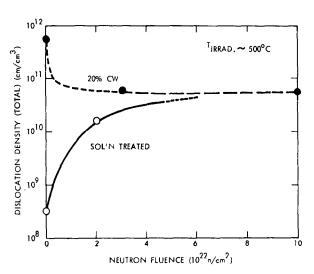


Fig. 15. Development of the dislocation density during neutron irradiation in type AISI 316 SS [82].

tron exposure. For AISI 316 this saturation density amounts to about  $6 \times 10^{10}$  cm/cm<sup>3</sup> at 500°C. As reported, this saturation level for the total dislocation density is independent of the displacement rate and the irradiation temperatures. The available data on AISI 316 20% CW indicate that for very high neutron fluences the network dislocation will not be changed by applied stresses. However, one has again to take into account that the actual dislocation density represents a dynamic equilibrium between the production of dislocations and their recovery. Hence such a parameter only gives information on the actual status and no insight into the development at lower neutron fluences.

Possible effects of stress on void formation do not necessarily result in an anisotropic growth of single voids as it is the case for the planar Frank loops. However, it is of principle importance to know whether swelling will be influenced by external stresses, since this would contribute to an additional isotropic dimensional change. Since most of the experiments do not allow an intermediate examination by means of destructive investigations the determination of the true irradiation-creep strains  $\varepsilon_{\rm IPC} = \varepsilon_{\rm TOT} - \varepsilon_{\rm S}$  are not very precise for intermediate data points.

Garner et al. [83] pointed out that there is evidence for an accelerated swelling in the presence of external stresses. Again most data are available for type AISI 316. Bates and Gilbert [84] showed by measuring the density of highly irradiated pressurized tubes that swelling increases for both cold-worked and solution-annealed specimen (fig. 16a). For comparison, recent results of type 1.4981 [73] in a 14% cold-worked pre-

treatment are given in fig. 16b. Here changes in diameter as well in length of the pressurized capsules were determined. In addition density measurements were performed.

The data for high fluence levels in fig. 16 indicate a linear stress dependence of swelling,

$$S = S_0(1 + B(T)\sigma_{Hv}),$$

where S and  $S_0$  are the total, respectively stress-free swelling,  $\sigma_{\rm Hy}$  the hydrostatic stress component and B(T) a possible temperature-dependent materials constant. For both materials in fig. 16 the material constants B(T) are in the same order of magnitude (table 3). Boutard et al. [85] report a weaker stress-dependence, as also indicated in table 3.

At the moment there is nearly no information about the temperature dependence of B(T). However, the few data on type 1.4981 indicate that the value increases by a factor of 2-3 when the irradiation temperature is

Table 3 Influence of stress on swelling for different austenitic stainless steels  $[S = S_0(1 + B(T)\sigma_{Hy})]$ 

Materials	Condition	Temp.	Fluence (dpa)	B(T)	Ref.
AISI 316	SA	475	42	4.1×10 <sup>-3</sup>	[84]
AISI 316	CW	477	44	$6.2 \times 10^{-3}$	[84]
Type 1.4981	CW	480	65	$4.5 \times 10^{-3}$	[73]
Type 316	SA			$1.2 \sim 1.8 \times 10^{-3}$	[85]
Type 316	CW			$1.6 - 3.8 \times 10^{-3}$	[85]

SA: stress-annealed; CW: cold-worked.

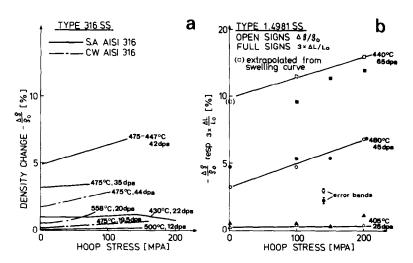


Fig. 16. Density changes and length changes of pressurized and unpressurized creep capsules: (a) type AISI 316 SS [84], (b) type 1.4981 SS [73].

increased from 440 to 480°C. Also the data on type 304 L [75] indicate a steep increase of this parameter with increasing irradiation temperature. Two mechanisms have been discussed so far [82] to explain the effect of stress on swelling:

- (i) increase of the reemission rate of vacancies from dislocations by hydrostatic stresses;
- (ii) change of the bias factors of voids by external stresses [86].

Wolfer et al. [87] have shown that the introduction of an enhanced reemission rate of vacancies from dislocations does not alter the nucleation rate of voids in stainless steels below 600°C. However, the change of the bias factors of voids by external stresses leads—when being introduced into the void nucleation theories [88,89]—to increased void nucleation rates, in accordance with observed increases in void number densities [81] of stressed samples.

It has been suggested that external stresses can affect both void nucleation as well as void growth [90]. The data presented in fig. 16 do not allow a decision between these two possibilities. In the case of type 1.4981 material the TEM investigations confirm the increased swelling, which is mainly caused by different size distributions of voids. However, it is not possible to distinguish between a stress-enhanced void growth, or the enhanced onset of void formation, since the development of the void structure could not be followed by TEM.

Very recent results on AISI 316 [89,91], where the development of volume changes was followed as a function of neutron dose, imply that the stress acts mainly by reducing the incubation dose instead by increasing the void-growth rate (fig. 17). This conclusion drawn from the experimental results is in accordance with an

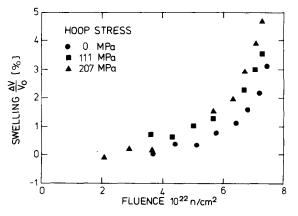


Fig. 17. The effect of stress on the swelling in type AISI 316 heat-treated [83].

earlier observation [92] that, once a stress had exerted its influence on swelling by reducing the incubation time, a subsequent variation of the stress level has no further influence on the steady state void growth.

In summary, these data show an increase of swelling with stress. The enhancement, however, does not only depend upon the stress level, but also on the irradiation temperature and on the swelling already achieved. Only for AISI 316 there seem to be enough data to warrant the conclusion that an enhanced void nucleation is the dominant physical mechanism. However, important questions, e.g. the influence of stress on the saturation level of void density or the effect, different stress states have upon the enhancement of swelling, are not yet answered.

#### 5. Swelling-creep interrelation

As was discussed already there is ample evidence that for most of the investigated austenitic stainless steels, irradiation creep rates increase with increasing neutron fluences. Several mechanisms could induce such changes, namely the development of the dislocations in the course of irradiation or changes in the bias factors for the different types of sinks, especially for loops, network dislocations or even voids, or microchemical changes, in the matrix which arise from the formation of precipitates and finally a possible swelling-creep interrelation.

Wolfer [25] has recently published a comparison between measured irradiation creep deformations in type AISI 316 and expected values calculated according to the SIPA model (fig. 18). These calculations are essentially based upon the microstructural data, especially the development of the loop and dislocation structure [81].

The comparison in fig. 18 indicates that the SIPA mechanism can account for a substantial fraction of irradiation creep deformation at moderate neutron fluences and for  $T > 450^{\circ}$ C. However, the model underestimates the experimental data at low doses. This gives rise to the assumption that glide processes also contribute to irradiation creep, especially if the dislocation density is low. The model predicts a constant creep rate at high neutron fluences since the dislocation density saturates already at moderate neutron exposures (-30 dpa). Therefore it would be necessary to introduce further parameters to account for a steadily increasing deformation rate.

One possibility for overcoming this problem is to correlate swelling and irradiation creep. Several of the

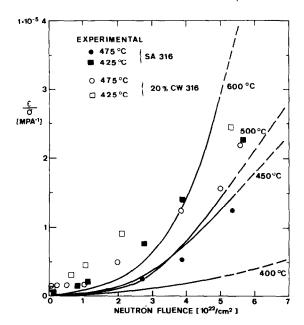


Fig. 18. Comparison of predicted and measured irradiation creep strain for type AISI 316 SS [25].

theories already formulated such a coupling. It is caused by the consideration that the net interstitial flux into dislocations corresponds to the net vacancy flux into the voids. The first suggestions for such a correlation between swelling and creep were made by Foster et al. [93], on the basis of results of residual stress measurements on fuel pin claddings of type AISI 304 SS [94] and on experiments with pressurized tubes [75]. From these data Foster et al. derived the following empirical equation:

$$\bar{\varepsilon}/\bar{\sigma} = K\bar{\phi} t + D\varepsilon_{c}, \tag{9}$$

where  $\phi t$  is the neutron fluence in dpa,  $\varepsilon_s(T,\phi t)$  the "linear" swelling and  $\overline{K}$  and D are material coefficients. For solution-annealed type 304 L stainless steel the coefficients have the values  $8.5 \times 10^{-7}$  [dpa<sup>-1</sup>] for  $\overline{K}$ , respectively  $1 \times 10^{-2}$  [MPa<sup>-1</sup>] for D.

Data published earlier on the creep deformation of pressurized tubes of type 1.4981 CW stainless steel [72] also suggested that irradiation creep accelerates when swelling starts (fig. 8). A recent analysis of the data [73] gives a similar relation to that found for AISI 304 in eq. (9). The coefficients  $\overline{K}$  and D found for type 1.4981 are:  $1.4 \times 10^{-6}$  [MPa<sup>-1</sup> dpa<sup>-1</sup>], respectively  $8 \times 10^{-3}$  [dpa<sup>-1</sup>].

The remarkably good agreement for the two steels

indicates that the mechanism for a coupling of swelling and irradiation creep is identical.

Such an interrelation has however important consequences: If the second term in eq. (9) enters with "linear" swelling rates in the order of 0.01% per dpa it would also dominate the irradiation creep. It is known from many publications that the so-called steady-state swelling rates for typical austenitic stainless steels have the order of 0.05%/dpa or more. Hence, one would expect that after the onset of swelling the irradiation creep should also show a stronger temperature dependence. Unfortunately the data basis is at present not sufficient to test this assumption over a broad temperature/fluence region.

A further consequence is an apparent change of the stress dependence of irradiation creep, if the swelling itself depends upon stress. Indeed it was possible to explain apparent deviations from the linear stress dependence of IPC in type 1.4981 SS by this additional stress-induced swelling contribution [73]. Several attempts have been made to explain a coupling between swelling and irradiation creep. Most of these have already been discussed in section 2. In addition, Weiner and Boltax [95,96] developed a model based on the well-known rate equation approach which considers specifically the point defect absorption by different sinks (voids, dislocations, and intersitial as well vacancy clusters). They conclude that an enhanced point defect recombination at vacancy loops and a swelling enhanced interstitial loop growth are the reasons for a swelling-enhanced irradiation creep.

At present, the I-creep model of Gittus seems to gives the best description of the coupling between swelling and irradiation creep. If one introduces the yield-stress values of the austenitic stainless steels mentioned above for 500°C, the factor of proportionality in eq. (5) agrees well with the experimentally determined D-constants in eq. (9). This would imply that in the absence of swelling the unequal partitioning of point defects at dislocations governs irradiation creep whereas at the onset of swelling the net interstitial absorption at dislocations contributes the dominant part to IPC by the I-creep mechanism.

Finally, there exists another very interesting approach to the problem of an possible correlation between creep and swelling. As already described, there is evidence that the formation of specific precipitates (e.g. carbides) enhances both swelling rates and irradiation-creep rates. In such a case the underlying mechanism for the enhancement of both phenomena would be the same. However, there is at present no quantitative formulation of such an interrelation.

#### 6. Conclusions

There are few general statements which can be made when describing the creep behaviour of austenitic stainless steels under irradiation: In a temperature/stress region (300-550°C), where thermal creep can be neglected, irradiation creep is weakly temperature dependent. In addition, for stress levels of practical importance (<150 MPa) a linear stress dependence is observed. Most of the experiments show a linear correlation of creep rate with damage rate; however, it seems that this statement may no longer be valid at very low irradiation temperatures.

There is ample evidence that with increasing damage levels the irradiation creep rates increase also. Several approaches have been made to explain this: by changes of the dislocation structure and total dislocation densities during the irradiation, by correlating irradiation creep with the onset of swelling or by considering changes in chemistry which are due to the formation of precipitates or to segregation phenomena.

The first approach has been quantified by introducing the changes of dislocation densities as function of fluence (SIPA model). Since a saturation of dislocation densities appears already at moderate damage levels ( $\sim 30$  dpa) further enhancement of irradiation creep rates should have other causes, e.g. changes of bias factors.

The interrelation of irradiation creep and swelling is an appropriate means to explain an enhanced creep at the onset of swelling. Such a correlation term will dominate the expected SIPA creep if the usually observed swelling rates are introduced. It can be explained by a model proposed as I creep and describes the data in the right order of magnitude. The prediction of such a model is that the temperature-dependence of irradiation creep will then follow those of swelling. Unfortunately the data available do not yet allow to confirm or reject such a creep behaviour.

A third approach is based upon the assumption that some elements like C, Mo etc. reduce irradiation creep as long as they are in solid solution. The same hypothesis has been put forward to explain the influence of these elements on swelling. Indeed, the present data indicate that unfavourable pretreatments like thermal ageing enhance both swelling and creep. In addition, a few experiments with chemically modified materials let us expect such behaviour. However, a clear understanding, of the physical mechanism is not yet arrived.

Deviations from the linear stress dependence of irradiation creep can indicate the transition to different creep mechanism, e.g. from SIPA to PAG creep. This is

especially expected at high stress levels and low dislocation densities. However, it is also possible that stressenhanced swelling can produce an apparent deviation from linear stress dependence if an interrelation between swelling and irradiation creep exists.

In a temperature regime where thermal creep and irradiation creep compete, the picture of in-reactor deformation is rather complicated. The conclusion which can be drawn from very few experiments is that, at high stress levels, thermal creep dominates. Its stress dependence shows the characteristic high stress exponents n. At low stress levels an in-reactor deformation is observed which shows a linear stress dependence and varies with the damage rate. The reduction of ductility and hence time-to-rupture, however, indicates that the rupture mode is not changed by this high-temperature in-reactor deformation. The development of a model is complicated further by the fact that in this temperature region the microstructure of the material can well be changed by the irradiation itself.

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