Jül-Conf-45

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I C A N S - V

MEETING OF THE INTERNATIONAL COLLABORATION ON

ADVANCED NEUTRON SOURCES

June 22-26, 1981

Radiation Effects in Targets and Structural Components of a Spallation Neutron Source

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Abstract

The basic effects of radiation damage on mechanical properties of metals are reviewed with special consideration of the operation conditions of high power spallation neutron sources. The possibility to use data from fast reactor experiments is discussed. Guide lines to material selection for a spallation neutron source are given with emphasis on the implications from radiation damage.

I. Introduction

It is well known that irradiation by energetic particles changes the physical and mechanical properties of solids /1-3/. Unfortunately most of these changes have negative consequences on the behaviour of materials under the operation conditions for example of a nuclear reactor. At present, material problems must be considered to be the limiting factors of the life times of fast breeder and future fusion reactors, and are closely related to the question of safe and economic operation of these advanced reactors. High power advanced neutron sources will share some of these problems and will possibly encounter some now ones due to their special operation conditions. That means, to demonstrate the engineering and economic feasibility of a high power spallation neutron source (SNS) also the material problems must be solved.

As current designs predominantly consider metallic materials for components of a SNS, the present surview is confined to the behaviour of metals and alloys under irradiation.

No large-scale material testing program comparable to those for fast breeder or fusion reactors are envisaged for the SNS. Therefore material selection will be mostly based on existing data from the above programs. The major part of radiation damage in the SNS target area are caused by the proton beam (800 - 1100 MeV) and by the energetic part of the spallation neutron spectrum. Both types of irradiation cause atomic recoil spectra which are different from recoil spectra obtained in present reactors or in current simulation experiments employing charged particles. Therefore caution must be taken when presently available data are used to estimate the response of structural materials in a SNS environment

II. Basic effects of energetic particle irradiation in solids

II.1 Displacement damage

An energetic particle (energy E, mass m) which is absorbed in a target atom (mass M), transfers a kinetic energy

$$T_i = E\frac{mM}{(m+M)^2} \tag{1}$$

to the atom. This energy must be modified by the distribution of the recoil energies of emitted secondary particles from nuclear reactions. Elastic recoils, which only predominate at small particle energies, cause a spectrum of recoil energies of the primary knocked-on atom up to a maximum energy of 4 T_i. The primary atom transfers its energy to surrounding lattice atoms, initiating a so-called displacement cascade. The number of defects $N_d(T)$ produced in a cascade of energy T can be calculated by a standard procedure /4/ if appropriate parameters are used (for Refs. compare Ref /5/). Integrating N_d over the distribution $d\sigma(T)$ of recoil energies which is derived from scattering experiments or calculated from nuclear models, yields the so-called displacement cross section σ_d:

$$\sigma_d(E) = \int N_d(T)d\sigma(T) \tag{2}$$

The displacement dose K in units of displacements per atoms (dpa) for a certain particle fluence Φ is then given by:

$$K = \sigma_d \Phi \qquad (3)$$

Calculations for 800 MeV protons are published in Ref. /6/ and for spallation neutrons in Ref. /7/.

K describes only the total number of interstitials and vacancies produced, but does not reflect their spatial arrangement. Especially the size of a cascade may be important with respect to recombination and agglomeration processes. The size distribution of cascades produced by a certain particle is determined by the spectrum of recoil energies.

It is known that fast-fission neutrons produce a significant amount of damage only within a rather narrow range of recoil energies while spallation neutrons /7/ and probably also 800 MeV protons /6/ produce much broader recoil spectra. Broad recoil spectra are typical for light ion irradiations already in the 10 MeV range. The fact that for example the recoil spectrum of 800 MeV protons extends to much higher energies, presumably does not produce qualitatively new radiation damage effects. This may be deduced e.g. from the similarity of 14 MeV neutron damage and that of fast fission neutrons, /8/, and can be explained by the observation that displacement cascades above a certain energy split into smaller units (subcascades) /9/.

II.2 Transmutations

Nuclear reactions following inelastic collisions between the irradiation particle and a target nucleus have three consequences which are closely interrelated:

a) Production of light reaction products, preferentially H and He. Helium is supposed to be of special importance due to its very low solubility in metals. Therefore it tends to segregate to grain boundaries and eventually causes embrittlement.

b) Change of the chemical composition of the irradiated material. This may induce phase instabilies or precipitations with important effects on mechanical properties.

c) Radioactivity from instable daughter nuclei. Radioactivity is of great importance with regard to maintainance and final disposal of structural components.

These effects may vary strongly for different elements and even isotopes. Therefore they should be known for each isotope of a structural alloy.

In table I some of the basic radiation effect data of a conceptional high power SNS design are compared to values expected in fast fission and fusion power reactors. For the SNS-design Mo and Al were used as examples for stationary and rotating structures, respectively. The difference in damage parameters between these two columns are primarily not specific for these materials but

Reactor	Breeder [a)]	Fusion [a)]	SNS (Germany) [b)]	
Material	Steel	Steel	Mo stationary	Al rotating
displacement rate [dpa/year]	50	20	700	4
appm He/year	10	400	$6 \cdot 10^4$	200
appm He/dpa	0.2	20	90	60
temperature [°C]	300-600	300-500	800	50
stress level [MPa]	60-120	60-200	100	80
cycles/year	10	10^4-10^8 [c)]	$3 \cdot 10^9$	$1.6 \cdot 10^7$

a) Ref. /10/

b) Ref. /11/

c) The lower value is typical for a magnetic device (Tokamak) the upper value for intertial confinement (Laser fusion).

Table I   Estimated irradiation parameters in different environments

are due to the fact that rotating parts in this design are only about 0.5% of the total time exposed to the proton beam. Table I shows that displacement rates and He-production in a stationary part would be orders of magnitude above the highest values expected in both fission and fusion reactors. On the other hand the data for the rotating parts are similar to those in a fusion reactor. Temperatures and stress levels of the SNS-design are inconspicuous but the high cycling frequences of the particle fluxes in the SNS are partly comparable only to inertial confinement fusion devices. The cyclic nature of damage production must be considered as an outstanding and widely unexplored peculiarity of the SNS.

III. <u>Mechanical property changes under energetic particle irradiation</u>

The basic irradiation effects described in the last section have implications on a variety of material properties. As mechanical property changes are assumed to have the most severe consequences on the operation of a SNS, they will be discussed now in more detail.

III.1 Dimensional changes

While sputtering is no longer considered a serious problem for structural materials with respect to surface erosion under high energy light ion or neutron irradiation, significant dimensional changes may be caused by void swelling, irradiation growth and irradiation creep /12/.

a) Void swelling
Voids are three dimensional aggregates of vacancies which nucleate preferentially under the action of gaseous impurities (helium) and grow due to vacancy supersaturation which is caused by the stronger attraction of self interstitials to dislocations. Void swelling shows a strong temperature dependence (Fig. 1): At low temperatures the low mobility of the vacancies causes high stationary vacancy concentrations and therefore enhanced recombination with the interstitials. Only at temperatures around half the melting temperature when vacancies become more mobile, recombination is reduced and maximum swelling occurs. At still higher temperatures thermal vacancy emission from the void surfaces competes with void growth, causing reduction of swelling.

The peak swelling temperature is shifted upward with increasing displacement rate, as high stationary vacancy concentration - on the low temperature side of the peak - and void growth - on the other side - persist to higher temperatures (Fig. 1).

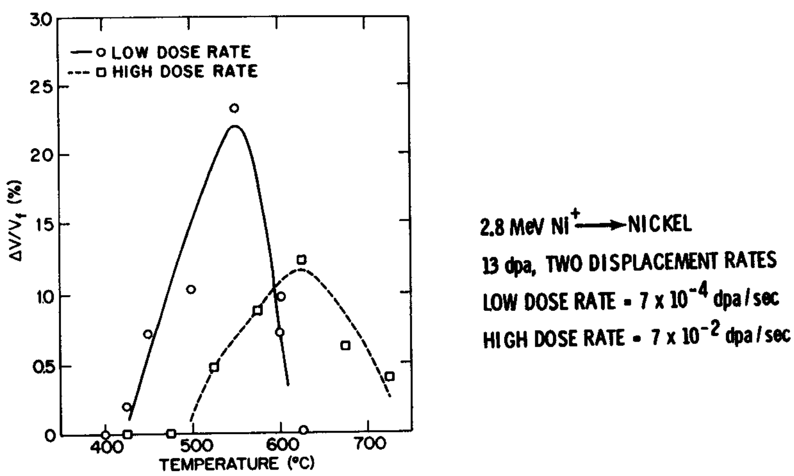

Fig. 1 Temperature dependence of swelling observed in Ni^{+} ion irradiation of nickel at two displacement rates /13/.

Void swelling is also affected by the dislocation density. While at very low dislocation densities swelling is suppressed by enhanced recombination, a very high dislocation density also reduces void formation by capturing vacancies before they are able to agglomerate. But tailoring of the microstructure is only of limited use to reduce void swelling or other property changes under irradiation, as under irradiation a new microstructure evolves which is largely independent of the starting conditions.

These considerations also apply to the dependence of void swelling on composition. It is well established that for example in stainless steel void swelling is reduced by increasing the content of nickel, silicon, carbon or other "light impurities". But microchemical changes due to segregation or precipitation occur under energetic particle irradiation already at displacement doses of a few dpa /14/ even in thermally stable alloys. That means, compositional modifications of an alloy may become ineffective due to microchemical changes already at rather low doses, while changes in overall chemical composition by transmutations occur usually much later.

The microchemical alterations during irradiation may also be the reason that swelling resistant alloys shows an incubation period for void formation of about the same dose range in which segregation occurs /14/.

Tensile stresses reduce this incubation period but seem to have no pronounced effect on the subsequent void growth /15/.

Theoretical calculations predict a general but moderate reduction of void swelling under pulsed irradiation compared to an equal steady state dose /16/. This is mainly due to enhanced recombination during the pulse-on time. At higher temperatures void shrinkage by vacancy emission during the 'off'-times further lowers the swelling rate.

A recent study of void swelling of aluminium under 800 MeV proton irradiation /17/ shows reasonable agreement with results from neutron irradiation.

b) Irradiation growth

In non cubic metals like Zirkonium alloys and uranium, a further type of dimensional change is observed which is called irradiation growth. The underlying mechanism is distinct from void swelling as no density change occurs, and distinct from irradiation creep as no external stresses are necessary. At the moment no exhaustive theory is available. In agreement with experimental evidence, current theoretical models attribute microscopic straining to the climb of dislocations and loops on the prismatic planes by the absorption of interstitials while the excess vacancies go to grain boundaries or at lower temperatures are also trapped at substitutional atoms. The models differ in their assumptions whether this separation of interstitials and vacancies is achieved by a preferential attraction of vacancies to grain boundaries /18/ or by preferential attraction of interstitials to dislocations /19/.

Macroscopic straining resulting from strains in the individual grains, depends on texture and probably grain size of the polycristalline materials. Similar to the case of void swelling at least part of the strain is recoverable by annealing to temperatures where vacancy emission occurs.

Irradiation growth is essentially linear in dose with some intermediate leveling off, observed for example in Zircaloy. It decreases with increasing temperature probably due to a reduced strength of the vacancy sinks.

No information on the effect of cyclic irradiation is available. As current theories predict enhanced straining under pulsed irradiation only in those cases where glissile motion of dislocations is involved, no large effect of pulsing on irradiation growth should be expected.

c) Irradiation creep

Irradiation creep is an irradiation induced, volume concerving plastic deformation under external stress. Unlike swelling or growth, creep deformation also occurs without irradiation and is a well known phenomena at temperatures above about half the melting temperature. This thermal creep is driven by thermally activated vacancies which interact with dislocations or at very high temperatures with grain boundaries. As the concentration of thermal vacancies drops exponentially with decreasing temperature, thermal creep ceases at temperature below about half the melting temperature. On the other hand the concentration of irradiation defects is essentially temperature independent. Therefore irradiation creep prevails below $T_m/2$ (Fig. 2).

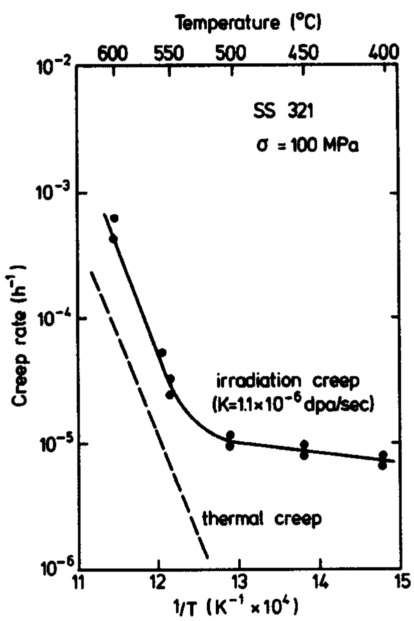

Fig. 2

Temperature dependence of thermal and irradiation creep in cold worked type 321 stainless steel /20/.

Several elaborate models for irradiation creep are proposed /18/. The most prominent are:

i) stress induced preferential nucleation of self-interstitial loops on those lattice plains which are oriented orthogonal to the external stress,

ii) stress induced preferential absorption of self-interstitials at such dislocations or loops, that the strain due to the incremental climb of the dislocation is parallel to the external stress,

iii) stress induced climb plus climb-enabled glide of dislocations. Irradiation defects enable the dislocation to climb across obstacles, while the major part of creep strain is achieved by glissile motion of dislocations.

At the time being, experimental results do not allow a safe decision for one of the above models. Some discrimination may be possible as the first two models predict a linear stress dependence of irradiation creep while the third model gives a higher stress exponent. As most metals investigated so far, show a transition from linear to quadratic stress dependence, a change of the prevailing creep mechanism seems to occur with increasing stress (Fig. 3).

In contrast to the incubation time observed for void swelling, irradiation creep is initially enhanced during some transient dose. Lateron irradiation creep strain is basically linear in dose as long as no significant alterations in microstructure and microchemistry occur. At high doses when void swelling is important an enhancement of irradiation creep is observed indicating a coupling of creep and swelling.

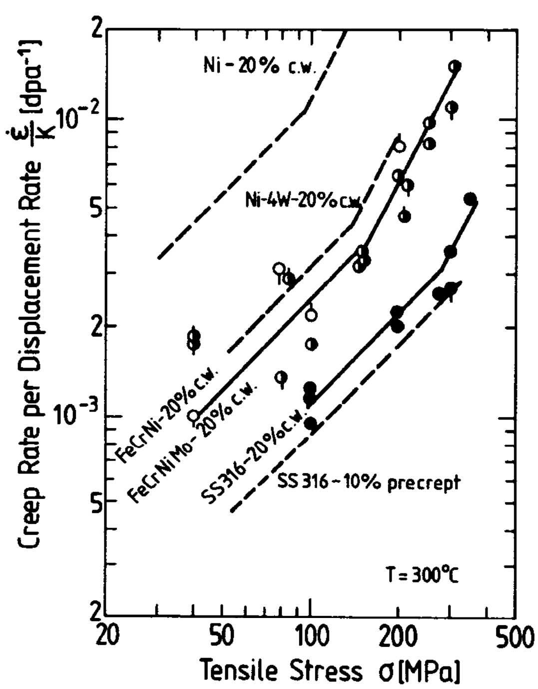

Fig. 3

Irradiation creep rates of 20% cold-worked Ni, Ni-4W, FeCrNi, FeCrNiMo, SS316, and of 10% precrept SS316 show linear stress dependence at low stresses and quadratic stress dependence at high stresses /21/.

The effect of cyclic irradiation on irradiation creep will strongly depend on the operative creep mechanism. For the first two mechanisms, the high defect concentration during pulse time will cause increased recombination. Therefore on the average a reduction in creep strain should be expected.
For the third model a strong enhancement of irradiation creep under pulsed conditions is predicted /22/: The basic idea is that under pulsed irradiation interstitials and vacancies arrive at different times at the dislocations due to their widely different diffusion constants. The separate arrival of only one defect species at a time enables enhanced climb to free the dislocation from obstacles. The subsequent glide motion separates the dislocation far enough from the obstacle, such that the following inverse climb motion under the action of the other defect species does not bring the dislocation back to the obstacle. A similar separation of interstitials and vacancies as under pulsed irradiation could be imagined under the action of very large cascades /23/ when defect distribution is very inhomogeneous. The expected enhancement of irradiation creep rate under pulsed conditions ranges up to factors of about 30, but depends strongly on pulse rate, temperature and material properties.

In conclusion a rough estimate should be given what dimensional changes must be expected. For technical alloys uniaxial elongation rates due to void swelling or irradiation growth range up to maximum values around 10^{-3} per dpa and to values slightly below 10^{-3} per dpa for irradiation creep (at 100 MPa). By appropriate choice of temperature the effects of swelling and growth may be reduced, while irradiation creep is almost independent of temperature.

III.2 Strength changes

Structural components are subject to external and internal stresses. Internal stresses may be caused by temperature gradients or inhomogeneous dimensional changes. Some beneficial effects may arise from irradiation creep which enables stress relaxation.

Strength is probably the only basic mechanical property where irradiation has an advantageous effect. A significant increase in yield strength (YS) and a smaller increase in ultimate tensile strength (UTS) is observed (Fig. 4) in most metals already at fairly low doses. This irradiation hardening is ascribed to dislocation pinning by irradiation induced obstacles, which were identified as dislocation loops in the case of pure fcc metals.

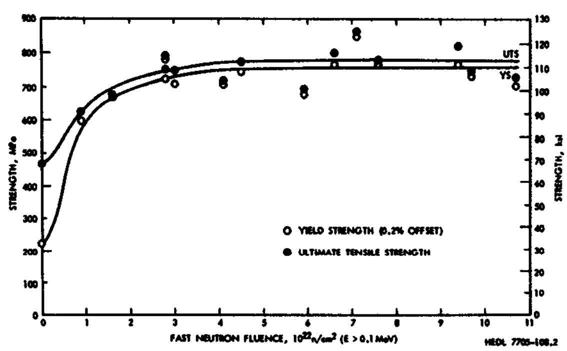

Fig. 4

Effect of neutron irradiation on the strength of annealed type 304 stainless steel at 370°C /24/.

In alloys with high interstitial impurity concentrations also radiation induced precipitation can contribute to obstacle formation. On the other hand in bcc metals occassionally the opposite effect of 'irradiation softening' is observed. This reduction of yield strength under irradiation is ascribed to a complex interaction of the dislocation structure with irradiation defects and alloying elements.

At elevated temperatures the strengthening effect disappears due to a reduced nucleation or even annealing of irradiation produced obstacles and due to an increased thermal mobility of dislocations (creep).

III.3 Ductility changes

Ductility is reduced under irradiation by different mechanisms mainly dependent on temperature and stress.

a) Low temperature embrittlement

The decreasing difference between UTS and YS (Fig. 4) with dose is accompanied by a decreasing elongation to fracture, that means with a loss in ductility (Fig. 5). Due to the close correlation of low temperature embrittlement with irradiation hardening, also the embrittlement disappears at

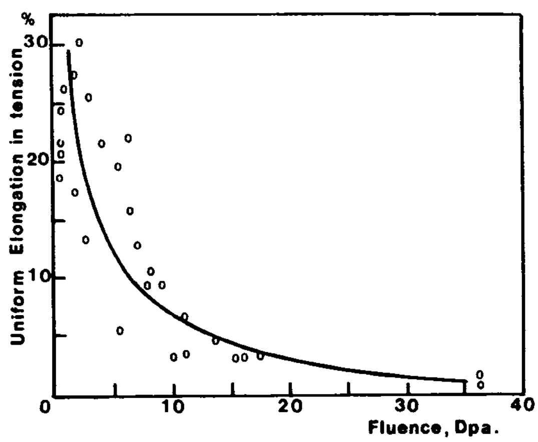

Fig. 5

Uniform elongation in tension of type 316 stainless steel at 400°C /25/.

elevated temperatures (above 450°C in stainless steels) by the reasons described above. Low temperature embrittlement is particular critical in alloys which undergo a ductile-to-brittle transition like most bcc metals and bcc alloys. In materials like ferritic steels the ductile-brittle transition temperature is raised by irradiation from below room temperature to a value within the operating range of e.g. reactor pressure vessels (Fig. 6). Spontaneous crack initiation and fracture is the possible consequence.

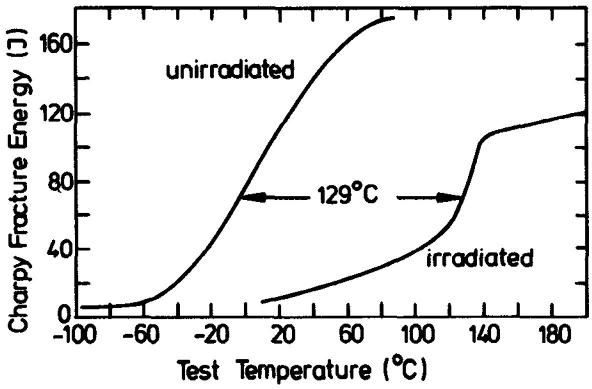

Fig. 6 Shift of ductile to brittle transition temperature of a Ni-Mo-Cr reactor pressure vessel steel irradiated to $1.25 \cdot 10^{20}$ fast neutrons per cm^2 (after Ref. /26/).

b) High temperature embrittlement

At elevated temperatures when low temperature embrittlement effects disappear new mechanisms of irradiation embrittlement become operative. Among these are irradiation-induced precipitation of new phases and void formation (see section III.1a). But it is now generally agreed that helium created by (n,α) processes (see section II.2) is the dominating factor.

'Helium-embrittlement' becomes noticeable already at concentrations of a few ppm (Fig. 7) and is accompagnied by intergranular fracture in contrast to transgranular fracture of helium free material. Elongation to fracture and rupture time are reduced and both depend strongly on temperature and stress. The application of a tensile stress drastically enhances the nucleation and growth of helium filled bubbles along grain boundaries. The grain boundaries eventually fail when a certain fraction of their areas is covered with bubbles.

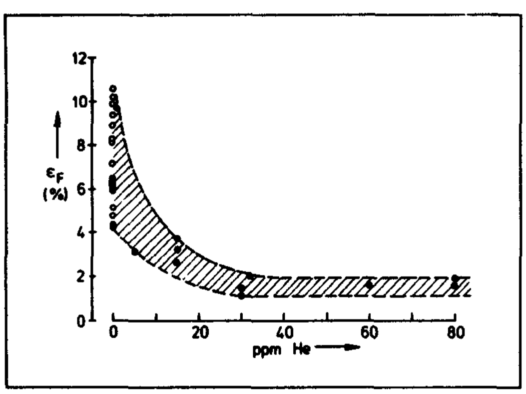

Fig. 7 Reduction of elongation to fracture under creep conditions of type 1.4970 stainless steel by implanted helium /27/.

It is not yet settled whether nucleation /28/ or growth /29/ of the bubbles is the time limiting factor for the embrittlement. So far, most information was obtained from post irradiation tests of neutron-irradiated or α-implanted specimens. Theory and recent experiments /30/ show shorter rupture times if the helium is produced in a specimen under tensile stress. On the other hand, simultaneous production of displacement damage may reduce embrittlement. This is documented by an investigation which is representative for SNS conditions: Irradiation of Al with 600 MeV-p /31/ indicates that under these irradiation conditions - high ratio of He to dpa production - more bubbles are formed near grain boundaries than for the lower He/dpa ratios obtained under fast neutron irradiation.

Only limited information on the effect of cyclic irradiation is available /32/, indicating enhanced coalescence of bubbles under pulsed conditions.

Helium-embrittlement is observed only at temperatures significantly above $T_m/2$. This fact and the strong dependence on temperature and stress may guide to reduce embrittlement by appropriate choice of parameters and materials /33/. Furtheron reduced grain sizes or finely dispersed precipitates - for example TiC in stainless steel /34/ - may be helpful.

c) Fatigue

Fatigue was soon recognized to be a serious problem in fast reactors. To reduce potential hazards, special consideration was given to unavoidable load ramps and shut down procedures. It is obvious that material response under this so-called low cycle fatigue condition which is characterized by low frequencies but high load changes may be entirely different from those in a SNS or an inertial confinement fusion system which both feature high frequencies but smaller stress changes.

Indeed opposite effects of irradiation on fatigue life are estimated in the low versus high cycle fatigue regimes (Fig. 8). In the low cycle fatigue regime strains are high and may even extend into the plastic regime. Therefore loss of ductility by the different mechanisms of irradiation embrittlement are supposed to reduce the fatigue life. On the other hand for high cycle fatigue, strains (must) remain well in the elastic regime. Therefore at not too high temperatures 'irradiation hardening' effects will increase fatigue life /35/.

Holding times under tensile stress during the cycles were found to reduce fatigue life by up to more than an order of magnitude /35/. This is explained in terms of creep-fatigue interaction, possibly with additional effects of the gaseous environment. Helium was also found to reduce fatigue life /36/. The largest effects occured under conditions which are typical for 'Helium-embrittlement' (see section III.3b).

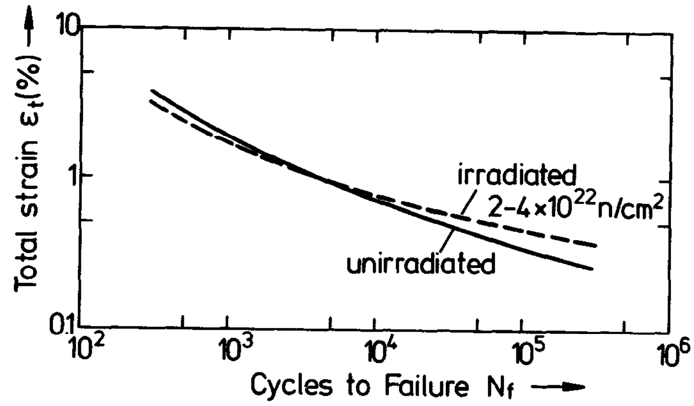

Fig. 8 Estimated fatigue properties of type 316 stainless steel after irradiation to a fast neutron dose of $2.4 \cdot 10^{22}/cm^2$. The results are based on tensile properties at 430°C /35/.

Inspite of the great importance which changes of fatigue properties may have on the economic and safe operation of advanced reactors (and SNSs), theoretical as well as experimental knowledge on this field is still very limited. Even without irradiation, fatigue is one of the least well understood mechanical properties. Furtheron experimental difficulties prevented in-pile tests up to now. Therefore the limited data which are so far available come from post-irradiation tests. From theoretical considerations /37/, simultaneous irradiation may have beneficial effects on crack growth, but only in a regime of temperature and dose rate, where appreciable irradiation hardening occurs during every load cycle.

IV. Guide line to SNS-material selection

The basic criteria for selecting materials for a high-powered SNS are similar to those for other large scala nuclear devices /38/. The following demands must be imposed on a candidate material:

1. Existence of a data base on mechanical and thermal properties and on irradiation effects.

2. Compatibility with coolants etc., corrosion resistance.

3. Fabricability and joining.

4. Adequate price and availability.

5. Low radioactivity (important for service and final disposal).

In the following those irradiation effects will be listed which probably impose the greatest problems to those SNS components which experience the most severe radiation damage:

1. Stationary windows (exposed to the pulsed, high energy proton beam): 'Helium-embrittlement' and probably 'high-cycle fatigue'.

2. Rotating windows, targets and cladding (exposed to the proton beam and to the spallation neutrons):
   'Fatigue' and 'irradiation creep'.
   For Al as cladding material also void swelling and He-embrittlement may be important.

3. Structure (exposed to spallation neutrons and moderated neutrons): 'Low temperature embrittlement'.

It is out of the scope of this paper to give a detailed recommendation for specific materials. Given the complex dependences of mechanical properties and irradiation effects on operation conditions, only for a well defined design the optimal materials can be selected (For a recent study compare Ref. /11/).

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