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Fusion Technology Institute University of Wisconsin 1500 Engineering Drive Madison, WI 53706

http://fti.neep.wisc.edu

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R. Bullough*, B.L. Eyre $^{\frac{\mathbf{I}}{2}}$ and G.L. Kulcinski $^{\mathbf{X}}$

ABSTRACT

To assess the suitability of a material for use as a core component in a fast reactor or for the first wall in a fusion reactor, it is necessary to know the irradiation damage behaviour of the material outside the usual materials testing data domain. In the present paper we propose a strategy based on a closely co-ordinated programme of experimental and theoretical research. The aim of this strategy is the systematic construction of a physically based model of the evolving damage structures. This would then allow both the necessary extrapolations of the data to the desired conditions to be achieved in a reliable fashion and provide a rational basis for the development of low swelling alloys for the two nuclear systems.

*Theoretical Physics Division

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¹Metallurgy Division AERE Harwell, Oxon, U.K.

XNuclear Engineering Department University of Wisconsin Madison, Wisconsin, U.S.A.

1. Introduction

In the last ten years considerable world-wide effort has gone into the search for low swelling alloys suitable for the core components of the fast reactor. After such a period and particularly since the emerging fusion reactor programme will also require similar low swelling first wall materials, it is appropriate to both review the previous effort and attempt to formulate a research strategy for the next decade that should avoid most of the shortcomings of the previous effort.

The choice of suitable materials for the structural components of a fast reactor core or for the first wall of a fusion reactor is constrained by the effects of irradiation on a potential materials dimensional and mechanical stability. In recent years, particularly since the discovery of the void swelling phenomenon in 1967, a great deal of research has gone into investigating the damage behaviour of a wide range of metals and alloys (1-5). Parallel efforts have been made to develop methods for analyzing and extrapolating the data, particularly to higher dose. These data analyses have proceeded along two main lines. The more widely accepted approach, which is being used to develop rules for predicting high dose reactor void swelling behaviour, is to subject the existing data to statistical analysis and thus obtain comparatively simple empirical relationships. Such an approach requires the input of only the void swelling as a function of dose, dose rate, stress and temperature and does not require any detailed knowledge or understanding of the damage structure. The second approach has been to construct theoretical models based on physical understanding which provide a framework for describing the evolution of the damage structure as a whole. While these latter models have been successful in identifying the important physical processes involved they are not ab initio theoretical models and require quantitative calibration using input based on accurate experimental observation of the damage structures particularly at low doses. Unfortunately, much of the experimental work on reactor materials has involved obtaining void swelling data without detailed structural observations using electron microscopy. Thus, it has not been possible to develop the physical models to the stage where they could be used to predict quantitatively the swelling behaviour of reactor components.

The purpose of the present paper is to suggest a combined strategy between the experimental and theoretical effort that would allow the experimental studies to provide relevant data for the physical model of the damage processes to develop. A theoretical framework can then be constructed that is able to provide both a feedback to the experimentalists and the means for reliable extrapolation of the observations outside the current data domain. The objective is thus not simply to gain a basic understanding of the damage mechanisms but to develop systematically the theory with the direct aid of relevant experimental data so that the suitability of a material can be accurately assessed. This programme is aimed directly at the void swelling and irradiation creep problems; it cannot alone lead to a quantitative physical model of irradiation embrittlement or fatigue failure. Such a development would necessitate also developing an understanding of the collective response to external stress of the dislocation component of the damage structure. However, we believe this latter understanding can only be achieved in the future if the evolution of the damage structure itself is first understood.

In the present paper, we first review briefly in section 2 the present situation to provide a background against which we present (in section 3) our suggested approach to the problem. Finally, some important conclusions are given in section 4.

2. Present Situation and Background

In this section we summarize the present situation with regard to the Fast Reactor core component and fusion reactor first wall materials investigations. The design requirements of the structural materials is first reviewed along with a discussion of the sources of relevant data and the method of analysis of the data, followed by an outline of the current approach to developing a void swelling resistant alloy. A short resume of the significance of the previous fundamental studies of irradiation damage is also given along with an outline of the present status of the physically based models of the damage phenomena. Lastly, we present a number of general conclusions which serve to introduce our suggested programme in section 3.

2.1 Design Requirements

Before one can intelligently devise a comprehensive programme to address the fundamental features of void swelling and creep in a nuclear environment the operating conditions must be clearly understood. These conditions are rather well defined for fission reactors but must, of necessity, be rather broadly stated for fusion reactors because the optimum approach to fusion (i.e., tokamaks, mirrors, inertially confined system, etc.) has not yet been chosen. A representative list of some of the important parameters that are associated with the two types of nuclear reactors are given in Table 1.

The operating temperature depends on the structure material which, in the case of the breeder, has been identified as an austenitic steel. Such a choice determines an upper limit of $\sim 600^{\circ}$ C and a lower limit (mainly due to the coolant) of $\sim 300^{\circ}$ C. The situation is much more complex for fusion reactors because there is no major feedback between the power producing reaction (D+T, D+D, or D-He3) and the structural material. Even with this increased flexibility, it is the opinion of most materials scientists that some type of austenitic stainless steel will be used in the near term ($\sim 1990-2010$) fusion reactors. This dictates a temperature range of $\sim 300-500^{\circ}$ C which is $\sim 100^{\circ}$ C

TABLE 1

Examples of Anticipated Structural Materials Requirements

For Fission and Fusion Reactors

Inertially Confined Fusion	300-500 (steel) 500-1000 (refract.)	10-10	10-30	200-500 (steel) 25-150 (refract.)	10'-10 ⁹	100-200	Reactor Life 300-1000 6000-20,000 (800-5000 refract.)	<10 <1
Magnetic Confined Fusion	300-500 (steel) 500-1000 (refract.)	3-10x10 ⁻⁷ (mirrors & tokamaks) 1-10x10 ⁻⁵ (theta pinch)	10-30	200-600 (steel) 25-150 (refract.)	~10 (Mirror) 103-105 (Tokamak) 3×10 ⁶ (0 pinch)	60-120	Acceptable >20	<5-10 <1
Fission Breeder (Steel)	300-600	9_0L°	∿50	∿10	010ء	60-120	, 100-150 20-30	5 < < >
Parameter	Temperature ^o C	Maximum Displacement Rate Instantaneous dpa s ⁻¹	Average ^x dpa y ⁻¹	Helium Gas Production appm y 1	Number of Power Cycles Per Year	Stress Level - MPa	Desired Lifetime Conditions dpa He (appm)	ΔV/V _o % (lifetime) Creep % (lifetime)

X70% PF

lower than for fission reactors because of the high helium production rate and helium embrittlement. If refractory metals (V, Nb and Mo) could be used, the maximum operating temperature could be increased above 500° C to $\sim 800^{\circ}$ C for V alloys and 1000° C for Mo and Nb alloys. Other alloys which have been proposed include those of Al and Ti. However, the useful operating temperature of Al alloys is too low (< 200° C) and the helium production rate in a 14 MeV neutron environment is too high, 300--400 appm He/MW/m². Ti or Zr alloys allow higher temperature application, but some are not competible with liquid lithium, and in the case of Ti, are also subject to high helium production rates. Further work on the Ti and Zr alloy systems is needed before a more definitive decision can be made.

The displacement rates in fast breeder reactors (which are limited by power density considerations) are in the $\sim 10^{-6}$ dpa \sec^{-1} range in the centre of a core. The displacement rates in a fusion reactor are not directly associated with the power directly in the plasma, but depend more on geometrical and economical considerations. Most reactor designs reach a reasonable compromise that lies in the 1-3 MW/m² range which translates into a $3\text{-}10\text{x}10^{-7}$ dpa for tokamaks and mirrors and $\sim 10^{-5}$ to 10^{-4} dpa \sec^{-1} for theta pinches. Inertially confined fusion reactors represent a much different case. The short burn time ($\sim 10^{-9}$ sec) and Doppler broadening result in ~ 10 dpa \sec^{-1} for ~ 10 ns pulse. At ~ 100 MJ per pulse, approximately 3-10 pulses per second would be required in economical systems and this amounts to $\sim 10\text{-}30$ dpa y^{-1} .

Once the dpa rate is determined, the amount of helium produced is a function of the neutron spectrum. This approach means that for steel, $\sim \! 10$ appm of He/year is produced in the centre of a breeder core and 200-600 per year in fusion. The use of refractory metals can lower this by factors of 4 to 8.

The number of times that the power level is cycled per year ranges from $\sim 10~\text{y}^{-1}$ in mirrors and fission breeders (because of normal power plant component failures) to 1000-100,000 per year for tokamaks (depending on impurity build up and available core flux) and $\sim 3,000,000$ per year for the theta pinch. Inertially confined systems experience from 10^7 to 10^9 "power" cycles per year. These latter numbers could be very difficult to design for because of the high stress or strain levels anticipated in inertially confined systems.

The stress levels are quite system dependent, but in the case of fission, they are dominated by the power density level and fuel expansion while in fusion, they are mainly associated with the high surface heat loading. The higher stress levels in laser systems are due to the pressure waves that could be generated by the various particle fluxes irradiated on the first wall.

Finally, one needs an idea about the desired (or at least acceptable) damage levels that the structure must withstand. In the case of fission, the design limits are set by the fuel burn up and neutron fluences of $2\text{-}3\text{x}10^{23}$ n/cm² (E >0.1 MeV) are required. This fluence translates into 100-150 dpa and 20-30 appm He in steel. Because of the close tolerances, swelling levels of < 5% must be maintained to prevent bowing and core compaction. Creep rates of < 1% during the lifetime would also be desirable to minimize the difficulties for insertion and withdrawal of fuel elements and control rods.

The situation for fusion is again, much more ill defined and complicated because it would be desirable if the structural metal lasted the full lifetime of a reactor (~ 30 years). However, it is unreasonable to expect any material to function after 300-1000 dpa and containing many thousand appm helium. Allowable swelling and creep values are not easily determined at this

time for fusion, but they should be of the same order of magnitude as for fission reactors.

It is intended that the values in Table 1 should be representative of the desired performance of nuclear structural materials, but not interpreted as ultimate requirements, especially for fusion. However, they are reasonable targets for commercial systems and if materials can be developed to fulfil these values, then it makes a commercial system much more feasible.

2.2 Prime Sources of Data

These consist of:

- (1) <u>Fast Fission Reactor Irradiations</u> using either rig irradiations of mainly <u>unstressed</u> specimens particularly in EBR II and DFR, or specimens obtained from experimental fuel element sub-assemblies. A limited number of steel specimens have been irradiated in high flux thermal reactors to produce large amounts (several thousand ppm) of helium in Nickel containing alloys. Some very low dose data ($\sim 10^{17}$ n cm⁻²) has been obtained from 14 MeV neutron sources on small specimens, irradiated at RT.
- (2) <u>Simulation Experiments</u> involving electron irradiation in high voltage electron microscopes (HVEM), irradiation with heavy ions or light particles using accelerators, and a limited amount of small specimen irradiation in neutron stripping facilities.

2.3 The Analysis of Data

In the more widely adopted procedure of mathematical modelling, the data is analyzed by using it to determine a series of parameters in an empirical functional representation of the data. The object of such modelling is

(1) to provide a convenient (easily computable) representation of the available data and thereby provide codes to guide the design of reactor components,

- (2) to facilitate extrapolation of reactor data outside the data base,
- (3) to provide a method for using simulation data for the prediction of in reactor behaviour of the same material.

There are several weaknesses in this procedure which can be summarized as follows:

- (1) Purely mathematical models take no account of physical mechanisms (and of course changes in physical mechanisms) and for this reason have not provided a successful method for using simulation data to predict in reactor behaviour. For the same reason they cannot be used to extrapolate the reactor data (dose, temperature, neutron energy spectra) with any confidence.
- (2) The mathematical models depend on the validity of the data and this presents a major difficulty in dealing with much of the reactor data for which the irradiation conditions are frequently not well defined. Moreover, a systematic approach has not been widely adopted in carrying out the reactor (and to a large extent the simulation) experiments and thus the data is often somewhat arbitrary.

It is both interesting and important to note that a parallel exists in the case of time dependent deformation behaviour and in particular, creep. It is now well established that creep mechanisms are a sensitive function of deformation conditions, i.e., time, temperature, substructure and stress system. Thus, it is not possible to predict the creep behaviour under a given set of conditions from test results obtained under entirely different conditions (usually short time, high strain rate and temperature) without any knowledge of mechanisms. This realization is the fundamental basis of Ashby's successful deformation map concept. It is the recognition that, as in the deformation map construction, the basic mechanisms prevailing in the irradiation situation must be identified and physically modelled before extrapolation is possible that underlies the strategy of section 3.

. 2.4 Development of Swelling Resistant Alloys

An important component of the Fast Reactor Programme has been the search for a void resistant alloy and this will also be a major feature of any DT fusion reactor programme. The source of data is again simulation experiments and limited reactor irradiation together with the use of empirical models for extrapolating the data base to the relevant reactor conditions. Two major difficulties with this aspect of the work can be identified

- (1) Extrapolation methods based on empirical analysis are invalid for the reasons already outlined.
- (2) Identification of swelling resistant alloys from a limited data base without any understanding of the physical reasons for the resistance could be dangerous. In any case we would suggest that such a procedure involves intuitive guess work which can be inefficient.

It is necessary to recognise the very real possibility that the result of this alloy development procedure could be the discovery of an alloy that is void resistant in accelerators or in the HVEM but which shows completely different behaviour under in service reactor conditions.

2.5 Fundamental Experimental Studies of Radiation Damage in Metals

A considerable amount of effort has been directed to achieving a fundamental understanding of irradiation produced defects in crystals over the last 20-25 years. Much of this work involved following the recovery of physical property changes with the aim of determining the transport properties of intrinsic point defects i.e., vacancies and self interstitials in crystals and irradiation was used as a convenient method of generating such defects. The irradiations were, in general, carried out at low temperature, e.g. $<20^{\circ}$ K, with various particles, e.g. γ , e⁻, various ions, and reactor neutrons. Although some insight has been gained into the diffusion properties of point

defects, the work has proved to be of limited value in understanding irradiation damage in commercial alloys used for the construction of reactors. The more obvious reasons for this are:

- (1) Because the parameters measured are related <u>indirectly</u> to the defects being studied there are fundamental difficulties in analyzing the data and considerable controversy still exists regarding their interpretation.
- (2) The experiments were mostly carried out on <u>pure</u> fcc metals having little direct relevance to reactor technology.
- (3) The irradiation conditions studied were <u>far removed</u> from the conditions under which reactor components operate.
- (4) The results give little insight into the fate of point defects not annihilated by recombination. It is this "surviving" population of defects which contribute to the structural changes, such as by aggregating to form clusters, migration to dislocations and grain boundaries etc., that cause the technologically important property changes in reactor components.

In recent years more emphasis has been placed on studying the configuration of point defects and their aggregation to form clusters. Use of X-ray scattering, Mossbauer and channelling measurements have enabled the configuration of self interstitial point defects and their trapping by substitutional impurities to be studied. These aspects of point defect behaviour are of fundamental importance to the formation and growth of point defect clusters. FIM studies of heavy ion and neutron irradiated metals are also proving to be of value, particularly in providing an insight into point defect distributions in uncollapsed displacement cascades.

The most direct contribution to our understanding of point defect clusters in irradiated crystals has however come from TEM studies of irradiated metals. A sound fundamental basis has been established for interpreting image contrast from extended defects. Moreover, the experiments have mostly

been carried out under conditions more closely related to those which reactor components will experience in service. The results from this work have led to important advances in our understanding of cluster morphologies and how their distribution depends on damage processes. Thus the results have exposed the important role played by recoil energy in governing the development of irradiation damage structures. It is worth emphasizing that these developments are already contributing to the interpretation of observed structural changes in commercial alloys irradiated under practically relevant conditions in reactor and in simulation experiments.

2.6 Physical Models of the Damage Process

Various physical models of swelling and irradiation creep have evolved over the last few years which have helped to indicate the many and varied physical and environmental parameters that can influence these phenomena. In particular, the use of rate theory has enabled the several mechanisms of swelling and irradiation creep to be identified and related to the prevailing irradiation conditions. However, there are basic parameters in the models that can, as yet, only be determined by direct experimental measurement. Furthermore, since there are often many competing physical processes prevailing, only relevant observation can determine the appropriate dominant process for the particular material and condition. However, the state of the theoretical understanding is such that we can define the experimental observations required for the construction of an appropriate physical model. It is this careful development of the physical model in complete and continuous coordination with appropriate (TEM) observations that forms the corner stone of our programme in section 3.

2.7 General Conclusions

(1) The unsystematic collection of limited data followed by empirical modelling is dangerous and likely to prove wasteful.

(2) The intention is to develop a relevant physical understanding of irradiation damage structures and thereby (a) establish a sound theoretical framework for predicting damage structures, (b) enable simulation data to be used for predicting reactor behaviour; and (c) provide a basis for identifying the necessary components for a damage resistant alloy.

3. Proposed Approach

Transmission electron microscopy studies have shown that irradiation damage structures consist of several distributions of different extended defect types which together make up the total damage structure. It is this resolution which provides a convenient framework for constructing a quantitative physical model of the evolving damage structure and of damage sensitive phenomena such as swelling and irradiation creep. In their present state of development, the models are best calibrated from a knowledge of the geometries and distribution (concentration and size) of the constituent extended defects as a function of dose over a limited range. In pure metals and single phase alloys, these defect components can be defined as follows:

- (1) The interstitial loop concentration N_{IL} and the irradiation produced dislocation density ρ_{IL} (the total length of interstitial loop perimenter per unit volume).
- (2) The total dislocation density $\rho_{IL} + \rho_N + \rho_g$. Here ρ_N is the network dislocation density that has either been introduced by prior cold work or has partly evolved from the interactions of growing interstitial dislocation loops and ρ_g is any "grown in" dislocation density (dislocations that show no ability to climb under the irradiation).
- (3) The cavity concentration C_c , average cavity radius r_c and the cavity size distribution.
- (4) The vacancy dislocation loop concentration N_{VL} . Such loops are not a feature of electron irradiation but are important for the calibration of recoil spectra effects.

The chemical rate theory of swelling and irradiation creep establishes the physical interrelation between the changing interstitial loops, voids, and vacancy loops and the other sinks for point defects in the body. For a given pure material, therefore, the above measurements at a series of temperatures

and dose rates can both calibrate the theoretical model and test its internal consistency to changes of the temperature and dose rate. On going to more complex multiphase alloys, it will be necessary to distinguish between phases having incoherent and coherent interfaces and to measure their distributions, in order to evaluate their comparative strengths as sinks for point defects. In these alloys it will also be necessary to attempt to quantify structural changes arising from resolution effects.

In the remainder of this section we outline in more detail the proposed programme. It comprises of a series of parallel experiments using electron irradiation in HVEM's, ion irradiation in accelerators, and neutron irradiation both in reactors and in 14 MeV neutron generators. This will then allow a systematic study to be made of the effects of changing dose rate and recoil energy on the damage structures. In the accelerator experiments the influence of both pre-injected and continuously injected inert gas are included, this being a particularly important factor in governing void swelling. The effect of stress will also be explored in the accelerator and neutron irradiation experiments in order to test critical aspects of the irradiation creep models. In all of the experimental work it is of the greatest importance that well characterized materials are studied and that accurate and complete evaluations be made of the damage structures.

The proposed programme can be basically divided into two main parts, although it is envisaged that work can proceed on both parts in parallel. The first part will consist of experiments on representative pure metals having fcc, bcc and cph crystal structures. The observations will be used to calibrate the fundamentally important parameters in the models, particularly those associated with the dislocation bias for self-interstitials and the material sensitivity to recoil energy. We consider it an essential first step to develop the models to a stage where they can reliably predict the behaviour

of pure metals. The second part of the programme is aimed at determining in a systematic way the influence of practically relevant metallurgical variables such as soluble and insoluble alloy (and impurity including H and He gas) additions, pre-irradiation dislocation structure, grain size, etc. on the damage structures generated by the different irradiation techniques. This will be used as a basis for parameterising the additional variables and incorporating them into the developing theoretical models which again must be continually tested for internal and predictive consistency.

3.1 Pure Metals

(a) Metals to be studied: fcc - Cu, Al, Ni bcc - α-Fe, V, Mo cph - Zr, Ti.

Of the fcc metals Cu is selected because a great deal is known about its physical properties and irradiation damage behaviour. Al has also been widely studied and because of its low atomic mass and comparatively high stacking fault energy, it presents a useful contrast to Cu particularly in terms of its sensitivity to recoil energy. Ni is of importance as a major alloy constituent in many of the alloys being considered for reactor components. The particular bcc metals were chosen because they represent different types of behaviour. Iron is, of course, a major constituent in ferritic and most austenitic alloys although relatively little work has been done on irradiation damage in α -Fe. V and Mo are refractory metals of particular interest as potential first wall materials in fusion reactors. They differ widely in their solubility for ' interstitial impurities and in mechanical behaviour. With regard to the cph metals, Zr was chosen because of its importance in water reactor core components. However, it is a difficult metal to study using TEM because of a tenacious surface oxide and thus Ti is also chosen as an additional cph metal because it is easier to study using this technique.

It is envisaged that all of the metals listed would be used in a high purity form and they should be thoroughly characterized in terms of impurity levels and starting structures.

(b) <u>Irradiation Experiments</u>

(i) HVEM: Irradiation with 1 MeV electrons results in essentially isolated Frenkel pair generation and thus effects arising from high energy recoils, such as displacement cascade production, will not play any role in determining the damage structure. An important objective of the experiments in the HVEM is to obtain the necessary data to calibrate the dislocation bias term and also to study interstitial loop and void nucleation. It is recommended that a majority of the experiments are carried out on He gas pre-injected specimens so as to obtain reproducibility. Some irradiation should also be carried out on He free specimens to confirm both the uniqueness of the bias term and to explore the influence of He on void and loop nucleation. The experiments will involve following both the dislocation (loops and network) and void components of the structure as a function of dose at a series of temperatures in the relevant range, i.e., $0.2 \rightarrow 0.6 \, T_{\rm m}$. It will be essential to place particular emphasis on thoroughly evaluating all components of the damage structures at low doses. This information is required to parameterise void and interstitial loop nucleation and to calibrate the bias terms. higher dose information can then be used for comparison with the theoretical predictions and thus enable the models to be thoroughly tested for internal consistency.

The model used for the analysis of the HVEM data does not require cascade effects and is thus relatively simple. The ability of such a model to represent the HVEM data in a consistent manner is thus the best indication that all the relevant physical factors for pure metals have been included in the model and the inclusion of cascade effects can then be confidently added.

(ii) Accelerator Irradiation: irradiation with heavy ions generates high energy primary knock-ons (typically 1-100 KeV) which, in turn, produce displacement cascades. Fundamental studies have shown that the vacancy rich centres can collapse to form vacancy loops and this, therefore, introduces a new component into the damage structure. The rate theory has been modified to take account of vacancy loop formation and it has been shown that this can have large effects on both void swelling and irradiation creep. However, the fundamental experimental studies have also shown that the probability of cascades collapsing to form vacancy loops is sensitive to a number of variables, including mass of the primary recoil and crystal structure of the target material. There is, therefore, a need to calibrate the cascade collapse efficiency for each of the metals as a function of irradiation temperature and this would be an important initial objective of the ion irradiation experiments. It will also be important to explore the influence of recoil energy on interstitial loop and void nucleation.

The ion irradiation experiments will mostly be carried out using selfions to avoid complications arising from the introduction of a second element. It is also proposed to use ions of \lessgtr 5 MeV so as to deposit the damage at least 1 \upmu below the incident surface. It will be both relevant and interesting to carry out also some irradiation with ions of very different masses in order to vary the recoil energy spectra. This could be important to understanding the damage structures in alloys containing elements of widely different mass numbers.

As in the case of the HVEM experiments, the procedure will be to carry out first a series of low dose experiments at a range of temperatures and to measure carefully the numbers and sizes of interstitial loops, vacancy loops, and voids. This will enable interstitial loop nucleation, void nucleation, and cascade collapse efficiency to be parameterised in the model.

Again high dose data will be compared with the predictions of the model, using the previously calibrated bias term, to test its validity in the new situation of high energy recoil production.

The heavy ion irradiation experiment will also be used to explore the effect of three further variables that are of particular importance to the behaviour of fusion reactor first wall materials. The first is concerned with the effect of inert gas and particularly the continuous production of He during irradiation. The experiments will involve a comparison of the damage structures in He free, He pre-injected, and in continuously He injected specimens. Thus, the experiments will require dual beam accelerator facilities. The results will again be analyzed to determine the influence of gas injection on loop and void nucleation, but the most important aspect will be to evaluate the effect of continuous He injection on void growth, particularly at high temperatures. The rate theory is already designed to take account of continuous gas production and the experiments will serve to test the validity of the theoretical predictions. The second variable is concerned with the influence of stress during irradiation. Here we are concerned in particular with the effect that stress has on interstitial loop nucleation and growth, which are both important aspects of irradiation creep models. The experiments will involve the irradiation of specimens to a range of low doses at different temperatures while being subjected to stress. The objective will be to determine quantitatively the effect stress has on the preferential nucleation of interstitial loops on planes that relieve the stress (i.e., those having the largest normal component along the stress axis) and to measure the growth rate of loops in different planes so as to calibrate the stress induced preference The effect of periodically varying the stress on a 0.1 to 1000 sec time scale will also be investigated. The third variable is concerned with the effect of pulsed irradiation on the damage structure. The experiments involve irradiation for short periods of time (<< 1 sec) and holding at temperature

between pulses during which recovery processes can occur. Careful evaluations will be made of all components of the damage structure as a function of the number of pulses, pulse time, and hold time at different temperatures. Again, the existing rate theory is able to deal with a pulsed irradiation situation and the experimental results will be used to test the validity of the model. Special pulsed experimental facilities will be required for this type of study.

(iii) Neutron Irradiation: A number of factors distinguish neutron irradiation in fission and plasma fusion reactors from electron or heavy ion irradiation in HVEM's and accelerators. First, the damage production rate is typically three to four orders of magnitude lower during reactor irradiation (although it is anticipated that the damage rates in laser fusion devices will be three to four orders of magnitude higher than those in accelerator irradiation for short periods of time. Second, the recoil energy spectra generated by fission and fusion neutron irradiations are in general harder than for heavy ion irradiation. Thus, a significantly higher fraction of the defects are generated in large displacement cascades. Third, He gas is generated continuously during irradiation at a rate that is a function of the neutron energy spectrum. An important objective of the neutron irradiation experiments is, therefore, to calibrate the effect these variables have on the damage structure in terms of interstitial loop and void nucleation and growth and cascade collapse efficiency. The procedure to be adopted will be the same as for the HVEM and ion irradiation experiments, namely, to obtain low dose data to calibrate the key parameters in the theoretical model and to use higher dose data to test the validity of the model. Specimens will be irradiated in the unstressed and stressed conditions and it is envisaged that most of the experiments will be carried out in different fission reactors (e.g., thermal and fast neutron test facilities) so as to vary neutron energy

spectra and He generation rates. Some irradiation in 14 MeV and spallation neutron generators would be included on a carefully selective basis (in view of the limited irradiation space) to explore the effect of high energy neutrons on critical aspects of the models, e.g. recoil energy sensitivity. In all of this work it will be essential to monitor the irradiation conditions and, as far as possible, control irradiation temperature and specimen stress levels using instrumented rigs.

3.2 Alloys

It is well established that void swelling and irradiation creep can be a sensitive function of a wide range of metallurgical variables related to composition and thermo-mechanical history. A dramatic example of this is the sensitivity of the swelling of austenitic Fe-Cr-Ni alloys to Ni composition. There is little quantitative, and in most cases little qualitative understanding of the role played by the metallurgical variables in terms of their influence on the different components of the damage structure. Thus, technological decisions to use alloys that are apparently resistant to void swelling or irradiation creep under accelerator or HVEM-irradiation conditions outside of the actual operating environment could prove to be dangerous.

An important objective of part II of the programme will be to develop a physical understanding of the role played by a range of metallurgical variables in governing the development of damage structure and to incorporate this into the theoretical models. The approach to be adopted will be similar to that described in part I, involving parallel electron, heavy ion and neutron irradiation of stressed and unstressed specimens followed by careful structural evaluation. Emphasis will be placed on carrying out the low dose irradiations required to calibrate the critical parameters in the model and these will be supplemented by selected high dose irradiations to test the predictive accuracy of the models. Clearly, the number of metallurgical variables that

could be investigated is very extensive and an exhaustive study would require an enormous programme. We have therefore attempted to select those variables which have technological relevance while at the same time allowing the basis of understanding established in part I to be extended in a systematic manner. Thus, the programme includes experiments on simple binary model alloys as well as on more complex alloys closer to the commercial materials used in practice. The variables to be studied can be conveniently categorized as follows:

- (i) Substitutional Solutes The main variables that should be studied are -
- (a) solute-atomic mass solutes having a significantly different mass to the matrix could modify the cascade size distribution.
- (b) solute-atom size over and under sized solutes could play an important role as point defect traps and by segregating to extended defects.
- (c) solute-induced changes in stacking fault energy this variable is particularly important in fcc metals (and possibly also in cph metals) and it could effect the morphology of planar clusters.
- (d) solute-induced changes in phase stability of particular interest are alloy compositions close to phase boundaries.

In order to explore systematically the effects of the above variables on damage structure, it is proposed to study the following alloy systems.

(a) Model Cu binary alloys containing Al, Zn, Si, Ge, Be, Ni. This series of elements would allow the atomic mass and size effect to be systematically studied. It is also known that all of the elements except Ni reduce the stacking fault energy from the value of 40 ergs cm $^{-2}$ for Cu down to as low as \sim 5 ergs cm $^{-2}$. Lastly, the solubility of these elements in Cu vary widely, ranging from complete solubility in the case of Ni to only a few percent in the case of Be.

- (b) Austenitic fcc alloys based on the Fe-Ni system in which Ni will first be varied systematically and then the effect of adding third substitutional solutes will be studied. The elements to be investigated will be those commonly used as alloy additives in austenitic alloys, starting with Cr and including Mo, Ti, Nb, V and Al.
- (c) Fe based ferritic alloys containing approximately 12% Cr but little or no Ni.
- (d) Refractory metal alloys based on Mo, and V with the addition of Ti, Zr and Cr.
- (ii) <u>Interstitial Solutes</u> This important class of solutes includes C and the so called residual gases, O, N, and H. In general, they are associated with comparatively large misfit strains, are soluble to a limited extent in most metals, and segregate to defects, interfaces, and surfaces causing significant changes in the interfacial and surface energies. Thus, in terms of irradiation damage structures, they may play an important role in the following respects:
 - (a) by acting as trapping centres for the intrinsic point defects,
- (b) by segregation to dislocations (loops and network) and thereby influencing the interaction of the dislocations with intrinsic point defects.
- (c) by segregation to the surfaces of cavities and by modifying the surface energy thus influencing both the nucleation and growth characteristics of the cavities.

The experimental programme will thus study the influence of C, O and N in solution on the different components of the damage structure. This will then be used to modify the theoretical models to take account of the effects. Emphasis will be placed on recognizing the effect C has on the dislocation component of the structure in austenitic alloys and bcc metals and in studying systematically the effect O and O have on the void component of the

structure in Cu, austenitic alloys and bcc metals.

- (iii) <u>Second Phase Particles</u> Second phase particles can be characterized in terms of two main variables which are to some extent inter-related.
- (a) Volume misfit this is important because of the long range interaction with point defects.
- (b) Coherence of interface this is important because it governs the behaviour of point defects when they reach the interface.

Precipitation also plays a major role in changing the solute composition of an alloy. Of particular importance in this context is the removal of the interstitial solutes from solution to form carbides, oxides, and nitrides. Clearly the range of precipitation phenomena that could be studied is very large. In the proposed programme we have attempted to select examples of the different types of behaviour. The main systems to be studied in the first instance are:

- (a) Cu containing coherent Cu-Co precipitates.

 Cu containing incoherent Si O₂ precipitates.
- (b) Al Cu alloys aged to varying degrees to produce G.P. zones, θ^{1} and Cu Al₂ precipitates.
- (c) Austenitic alloys (1) Around the 316 composition and containing $\sim\!\!0.05\%$ C to form semi-coherent $\rm M_{23}$ C_6.
 - (2) Containing \sim 0.1% C + Nb, Ti, or V and heat-treated to form misfitting incoherent carbides.
- (3) High Ni alloys containing Ti and Al to form coherent (Ni Ti) Al3.
- (4) High Ni alloys containing $\sim 0.1\%$ C and Cr, Ti, Nb and V to form different types of carbides.
 - (d) Refractory metals Mo, and V

- (1) containing only carbon to form the carbides $Mo_2 C$, and VC.
- (2) alloyed with Ti, Zr and Cr and carbon to form carbides with these elements.
- Dislocations: As well as the irradiation induced dislocation component of the damage structure there are the pre-existing dislocations which can be broadly classified into those grown-in and those introduced by deformation; i.e., the so-called cold work dislocations. In a complete physical model of the damage structure it is essential to include these pre-existing dislocations as intrinsic point defect sinks. It is, therefore, proposed that a systematic study be made of the configurational changes in these two types of dislocations in specimens irradiated to low doses. Although it will not be possible to control the grown-in dislocation structure, the deformation dislocation density and distribution can be varied in a systematic manner. In these experiments it will be important to study both pure metals and alloys to elucidate the role played by solute atoms and precipitate interactions with dislocations. Direct observation in the HVEM will be particularly valuable in providing a qualitative insight into how the pre-existing dislocation configurations are modified during irradiation. In addition, parallel irradiations using electrons, heavy ions, and neutrons will enable the influence of the pre-existing dislocations and the irradiation induced defect structure to be quantified and thus incorporated into the physical models.

4. Conclusions

The current understanding of damage processes and the kinetic theory of the evolution of the damage structures are both sufficiently developed for the combined strategy outlined in section 3 to be successful. The fundamental object of the suggested approach is to construct a quantitative and physically based model of swelling and irradiation creep which can be used for the reliable extrapolation of experimental data.

We consider the basic approach proposed here of carefully characterising all components of the damage structure in order to construct a comprehensive physical model of the evolving damage structure to be definitive. However, there is clearly considerable latitude on the choice of metallurgical parameters to be studied and these could well be dictated by other design requirements. We have attempted here to define a systematic approach to categorizing the different variables in terms of their effect on the damage structure.

The development of such physically based models will also give valuable insight and guidance to the interpretation of the obviously valuable damage data that is also obtained from the inspection of actual reactor components; the essential point being that such materials should also be subject to the same careful characterization of damage structure as suggested in the present programme to enable a correlation with the physical models to be made.

Finally, though we believe the suggested approach could significantly contribute to the design and selection of fast reactor core component materials, we believe it is absolutely essential for solving the corresponding fusion reactor material problem. The latter programme will rely for the foreseeable future, on data from simulation experiments and there is no way

such data can be used to predict behaviour of the material in the fusion reactor environment without an appropriate understanding of the underlying physical processes: the development of a physical model is thus essential.

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