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FUNDAMENTAL BEHAVIOR OF STRUCTURAL MATERIALS IN FAST REACTOR ENVIRONMENTS

T. T. Claudson, WADCO Corporation, Richland, Washington
 E. E. Sinclair, U. S. Atomic Energy Commission, Washington, D. C.
 B. R. T. Frost, Argonne National Laboratory, Argonne, Illinois
 A. Boltax, Westinghouse Electric Corporation, Pittsburgh, Pa.
 H. Pearlman, Atomics International, Canoga Park, California
 C. N. Spalaris, General Electric, San Jose, California
 J. R. Weir, Jr., Oak Ridge National Laboratory, Oak Ridge, Tenn.

I. INTRODUCTION

The successful design and operation of a Liquid Metal Fast Breeder Reactor (LMFBR) as an economical power plant depends greatly upon the development of nuclear materials technology. Since 1961 the U. S. Atomic Energy Commission has sponsored research and development programs related to (1) the understanding of the effects of nuclear reactor environments on behavior of structural and cladding materials and (2) providing sufficient data and information to permit safe and economical design of each major reactor system. In recent years these programs have been directed more at providing materials technology required for the LMFBR program. Specific areas of interest include (1) the effects of fast neutron irradiation on mechanical properties, (2) swelling and irradiation induced creep of cladding and structural materials, and (3) metallurgical structural stability of alloys. The major materials used in cladding and structural components of proposed LMFBR's are Types 304 and 316 stainless steel. Some consideration is being given the stabilized grades as Type 347 as well as Incoloy 800. Irradiations are performed in the EBR-II in the temperature range of 370 to 815°C with goal fluences exceeding 1×10^{23} n/cm² ($E > 0.1$ MeV). While the major approach is to obtain pre-and postirradiation data, efforts are underway using instrumented tests to determine creep properties in-reactor. Proton and heavy-ion accelerators are also being utilized to produce material damage to evaluate the effects of flux rate and high fluence levels on metal swelling and irradiation induced creep. This paper presents a survey of the present knowledge of the material technology in these areas of interest.

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II. MECHANICAL PROPERTIES OF IRRADIATED AUSTENITIC STAINLESS STEELS

Systematic studies of mechanical properties for irradiated austenitic stainless steel are now providing data required for design of fast reactor in-core components. This section of the report presents a summary of present test results pertaining to the effects of neutron irradiation and environment on the pre-post tensile, creep-rupture and biaxial stress rupture properties of austenitic stainless steel. Specimen neutron fluence levels were monitored and are generally considered correct to within $\pm 20\%$. Irradiation temperatures were calculated based upon EBR-II gamma heat levels, SiC temperature monitors or melt wires and are considered accurate to within $\pm 50^\circ\text{C}$. Recent data are presented which established the need for information related to the effects of controlled reactor sodium environments at temperatures above 593°C .

Tensile Properties - Among the austenitic stainless steels, the post-irradiation properties of Type 304 stainless steel have been the most thoroughly investigated. Carlander et al [1], performed tensile tests on Type 304 stainless steel components removed from EBR-II and recent results on Type 316 and a Ti bearing Type 304 stainless steel are available. The yield stress and elongations of Types 316, 304, and Ti modified Type 304 stainless steel are shown in Figures 1 and 2, respectively. At the irradiation temperatures of $450\text{--}460^\circ\text{C}$, the yield stresses rise rapidly with fluence and saturation appears to occur at a fluence of about $3 \times 10^{22} \text{ n/cm}^2$ ($E > 0.1 \text{ MeV}$). At 540°C saturation occurs at a lower fluence. There is some scatter in the elongation results; however, irradiation clearly has a pronounced effect on the ductility.

Although the data are meagre, several qualitative conclusions regarding the effects of fast neutron irradiations on the tensile properties of Type 316 stainless steel are consistent with the data on Type 304 stainless steel shown in Figures 1 and 2. The postirradiation yield stress is a stronger function of the irradiation temperature than the testing temperature; that is, the rapid rise in yield stress with fluence is very irradiation-temperature sensitive in both alloys. The ductility (elongation) for annealed Type 316 stainless steel is higher at 450°C than that of Type 304 stainless steel and is lower at 540°C . The strength remains higher and the ductility lower for cold worked Type 316 stainless steel than for the annealed alloys.

Tests have been completed which compared the effects of irradiation and environment on the tensile properties of Types 304, 316L, 316, 321, 347 stainless steel and Incoloy 800 each in the mill annealed and carbide agglomerated conditions [2]. The ductility, yield and ultimate strength, exhibited by all alloys are strain rate dependent. Results indicate that (a) stainless steels of wide alloy composition and structural differences exhibit a spectrum of ductilities which must be taken into account in a core performance analysis and (b) neutron exposures of $6 \times 10^{21} \text{ n/cm}^2$ ($E > 1.0 \text{ MeV}$) induced properties degradation equivalent to the degradation produced by exposure of unstabilized austenitic stainless in flowing sodium at 704°C for 5,000 hours. Most mechanical properties data available to date were determined from

specimens that were exposed either in inert atmospheres, stagnant sodium (capsules) or capsule bodies exposed to flowing sodium not exceeding 480°C.

Creep Properties - Postirradiation creep rupture tests have been conducted on solution treated Type 316 stainless steel and on solution treated Type 304 stainless steel [3,4]. Specimens were irradiated in EBR-II to a maximum fluence of about 7×10^{22} n/cm² ($E > 0.1$ MeV) over a temperature range of 370°C to 815°C and tested in the range 537 to 760°C. For both steels, specimens irradiated at temperatures below about 590°C there is a large decrease in the steady creep rate, primary creep and rupture strain as compared to specimens irradiated to the same fluence level but at higher temperatures. This is predominately due to the relatively larger amount of displacement damage, void production and dislocation structure produced at the lower irradiation temperature.

At higher irradiation temperature, Type 316 stainless steel specimens exhibited lower primary creep but approximately the same creep rate as control specimens. Some reduction in total rupture strain was observed (10% versus 26%) as well as a reduction in rupture life, 65 versus 375 hours. However, the most marked change was due to increasing the exposure level to 7×10^{22} n/cm² ($E > 0.1$ MeV). In this case the material was unable to withstand strain beyond primary creep at the 25,000 psi stress level. These data indicate that high temperature helium embrittlement is a major design factor for components operating under high stress at elevated temperatures.

Figure 3 summarizes rupture life data for annealed Type 316 and Type 304 stainless steel irradiated at temperatures indicated and tested at 650°C and 600°C, respectively. A reduction in rupture life has been observed at all irradiation temperature and fluence levels. The effects of irradiation and test temperature and neutron fluence on creep ductility are shown in Figure 4. The stress-rupture results appear to be consistent for the two steels. The lower irradiation temperatures produce larger effects on the rupture life and ductility.

The biaxial stress-rupture properties of mill-annealed Type 316 stainless steel have been determined following an irradiation at about 565°C to a fluence of 1.0×10^{22} n/cm² ($E > 0.1$ MeV) [5]. Using initial true stress as a basis for comparison, a reduction in rupture life of the Type 316 stainless steel was found to exist at all test temperatures. The reduction at 538, 649, and 760°C was found to be a factor of about 0.025, 0.50, and 0.10 of the unirradiated values, respectively, indicating a reduced effect with increasing test temperature. The creep rate data show higher average creep rates of irradiated material at a test temperature of 538°C. The average creep rates of irradiated specimens at 650°C are virtually the same as the unirradiated rates. Although the data at 760°C are quite limited, it appears that irradiated creep rate values are again higher than unirradiated values. The observations at 538 and 650°C are in agreement with uniaxial creep data for solution annealed Type 316 stainless steel from the same irradiation experiment.

FIGURE 1. THE EFFECT OF IRRADIATION ON THE YIELD STRESS OF TYPES 304 AND 316 STAINLESS STEEL WHEN TESTED AT THE IRRADIATION TEMPERATURE

FIGURE 2. THE EFFECT OF IRRADIATION ON THE DUCTILITY OF TYPES 304 AND 316 STAINLESS STEEL WHEN TESTED AT THE IRRADIATION TEMPERATURE

FIGURE 3. EFFECT OF FAST NEUTRON FLUENCE ON THE RUPTURE LIFE OF ANNEALED TYPE 316 STAINLESS STEEL AT 650°C (25,000 psi) AND ANNEALED TYPE 304 STAINLESS STEEL AT 600°C (27,000 psi)

FIGURE 4. CREEP DUCTILITY AT 538°C FOR ANNEALED TYPE 316 STAINLESS STEEL AND AT 600°C FOR TYPE 304 STAINLESS STEEL

Figure 5 shows a summary of data obtained from burst testing Type 304 stainless steel irradiated in the EBR-II and shows a marked decrease in ductility due to irradiation. The hoop strength was also found to increase with increasing fluence [2].

III. SWELLING AND CREEP OF AUSTENITIC STAINLESS STEEL

One of the more important design problems in LMFBR's is concerned with the dimensional stability and mechanical interaction of the subassemblies which constitute the core. To solve this problem it is necessary to know or predict the void swelling and effective creep or stress relaxation rates in a fast neutron environment as a function of temperature, fluence, flux and applied stress. Efforts include theoretical and empirical analysis, evaluation of components and specimens irradiated in the EBR-II and positive ion bombardment studies.

Theoretical - The importance of void formation to fast reactor technology has spurred the development of physically realistic models for void and loop nucleation and growth during irradiation. The approaches taken by Harkness and Li [6], Straalsund [7], Gulden and Kaae [8], Bullough, Eyre and Perrin [9] and Wiedersich [10] towards growth are similar and are based upon classical diffusion theory. All assume that void growth arises from a supersaturation of vacancies created by neutron irradiation. Void growth occurs when the net rate of vacancy flow to voids exceeds the net flow rate for interstitials. Since equal amounts of vacancies and interstitials are produced during irradiation and equal amounts are annihilated as a result of recombination, the excess vacancies that condense at voids must be balanced by an excess of interstitials condensing on other sinks such as dislocations and interstitial loops. Thus two requirements must be met to have void growth; (1) sinks other than voids take up the interstitials and (2) a driving force or bias tends to cause the vacancies to migrate to voids and the interstitials to other types of sinks. It is generally agreed that the most likely bias is a higher sink efficiency or capture cross section of dislocations and loops for interstitials than for vacancies due to stress assisted diffusion effects as considered by Ham [11].

While the growth portion of the phenomenon appears to be relatively well understood, nucleation poses a difficult problem in current theory. Consequently few attempts have been made to develop swelling models which incorporate both nucleation and growth. Bullough and Perrin considered that nucleation must eventually cease after some time, and make an extensive analysis of void growth assuming given void and dislocation densities. Consequently their model does not provide an accurate description of the swelling process at low fluences where sink densities are changing with exposure. These effects can be important in determining apparent growth rates and size distributions because the vacancy and interstitial supersaturations depend on sink density. Brager, et al [12] attempted to take these effects on growth into account by incorporating an

empirical estimate of the nucleation rates within their analysis. Harkness and Li were the first to advance a swelling model which combined a theoretically developed nucleation theory with classical growth theory in an attempt to describe the total phenomenon of swelling. In their analysis it was assumed that the nucleation behavior of voids in the presence of a vacancy supersaturation would be similar to that described by the classical nucleation theory of Becker and Doring [13]. The presence of interstitials was not considered in their analysis of nucleation. Katz, Wiedersich and Russell have recently extended homogeneous nucleation theory to take the presence of both types of defects into account. While considerably different in detail, their model shows the same general trends as the nucleation theory proposed by Harkness and Li: a sensitive dependence of nucleation rate upon point defect supersaturation. This particular feature causes models based on homogeneous nucleation to predict cessation of nucleation at a very early time during irradiation. Experimental data, however, suggest a more constant nucleation rate, which may be due to the influence of helium on the nucleation phenomenon in stainless steels.

Experimental - Investigators [14-24] have determined the density change of structural and cladding materials in order to (1) provide guidance as to the nucleation and growth kinetics of void formation for theoretical consideration and (2) to give statistically significant guidance to the designers of the FFTF and other LMFBR systems. Specimens have been examined from high fluence EBR-II components irradiated over a relatively narrow temperature range, 370-480°C. Specimens of Type 304 and Type 316 have also been irradiated to a maximum fluence of 5×10^{22} n/cm² at temperatures to 750°C to evaluate the effects of cold work and metallurgical structure on swelling behavior. In the absence of a well defined theoretical model describing swelling of austenitic stainless steel an analysis of all available data has resulted in empirical equations (1) and (2) relating stress free swelling of solution treated and 20% cold worked material, respectively, to fluence and irradiation temperature.

$$\% \Delta V/V = \phi t^{2.05-27/\theta + 78/\theta^2} [(T-40) \times 10^{-10}] \exp [-0.015(T)-5100/T + 32.6] \quad (1)$$

where ϕt = total neutron fluence $\times 10^{-22}$
 $\theta = T-623$
 $T = ^\circ K$

$$\% \Delta V/V = 9 \times 10^{-35} (\phi t)^{1.50} [4.028-3.712 \times 10^{-2}(T-273) + 1.0145 \times 10^{-4}(T-273)^2 - 7.879 \times 10^{-8}(T-273)^3] \quad (2)$$

ϕt = neutron fluence ($E > 0.1$ MeV)

Recent swelling data on solution treated, 27% and 75% cold worked Type 316 and 50% cold worked Type 304 stainless steel irradiated over a 370°C-760°C temperature range to a maximum fluence of 5×10^{22} n/cm² ($E > 0.1$ MeV) have been obtained [25]. These data

showed markedly the effect of cold work on the swelling behavior and composition of austenitic stainless steel in that little or no swelling was observed in the cold worked Type 316 stainless steel over the entire range of irradiation temperatures and fluence levels. The solution treated Type 316 material showed some swelling at temperatures to about 540°C but lower than would have been predicted by equation (1) above derived from Type 304 stainless steel data. The trends changed with increasing fluence for the solution treated Type 316 and 50% cold worked Type 304 stainless steel. Swelling in both these alloys increased with increasing fluence exhibiting a peak at about 425°C and dropping to near zero swelling at 540°C after exposures of 5×10^{22} n/cm² ($E > 0.1$ MeV). A second swelling peak not evident in Type 316 stainless steel was observed for 50% cold worked Type 304 stainless steel at 620°C. This second peak indicates a second controlling mechanism at the higher temperatures possibly associated with cold work recovery and subsequent nucleation and growth of voids on second phase particles.

Stainless cladding obtained from mixed oxide pins irradiated to 65,000 MWd/Te (4.0×10^{22} n/cm² ($E > 0.1$ MeV)) and 105,000 MWd/Te (7.4×10^{22} n/cm² ($E > 0.1$ MeV)) in EBR-II was measured to determine swelling. The experiment included both pellet and powder (U, Pu)O₂ fuel contained in 0.250 in. OD by 0.016 in. wall cladding of stainless steel Types 316 and 347. Volume increases of up to 8.7% after a neutron dose of 7.3×10^{22} n/cm² ($E > 0.1$ MeV) at 520°C were measured in Type 316 stainless steel and up to 10.0% for similar conditions in Type 347 stainless steel [26].

Transmission electron microscopy (TEM) showed that swelling does not occur in solution treated Type 304 stainless steel at irradiation temperatures in excess of 680°C. Very large (greater than 2000 Å diam.) voids were found in unstressed specimens irradiated at 500-640°C. A length of solution annealed Type 304 stainless steel strip (0.200 in. wide by 0.010 in. thick) was exposed in the EBR-II core region to temperatures in the range 530-680°C and fluences to 5.4×10^{22} n/cm² ($E > 0.1$ MeV). No voids could be located in specimens irradiated at ~ 680°C. The temperature dependence on void size and number density from this specimen and others is shown in Figure 6. Because the Type 304 strip was not under stress during irradiation, it was concluded from the void size data that an external stress system is not a prerequisite for the formation of very large voids. Figure 6 also shows that voids become larger in size and fewer in number as the irradiation temperature increases [27].

Effects of Stress on Stainless Steel Swelling - Analysis of the irradiation behavior of stainless steel indicates that stress can have a significant effect on the swelling of stainless steel over the temperature range of interest in fast reactors. A recent evaluation of this subject by F. Garner, et al [28] identified three mechanisms for stress effects on stainless steel swelling, including: (1) the growth of grain boundary cavities or helium bubbles under hydrostatic tension, (2) the hydrostatic component of irradiation creep and (3) the growth of voids under hydrostatic tension. The growth of grain

boundary cavities or helium bubbles under hydrostatic tension has been reported by numerous investigators indicating density decreases of 0.3% at diametral strains of 6% [29-33].

The earlier investigations of irradiation creep of stainless steel indicated that nonconservative volume changes are caused by the hydrostatic component of irradiation creep. The magnitude of this effect for hydrostatic tension has been predicted to be as high as 3.2% at 30,000 psi stress at 450°C for solution treated Type 316 stainless steel at a total fast fluence of 5×10^{22} n/cm². Hydrostatic compression would be expected to reduce the magnitude of swelling. Analysis of the results from mixed oxide fuel pins was performed with the OLYMPUS-II code [34,35]. The data are consistent with the thesis that the systematic increase in excess swelling for the fuel pins is caused by the increasing fuel smear density which in turn causes an increase in irradiation creep in the cladding. The volume change calculated from the hydrostatic tension portion of irradiation creep is consistent with the indicated excess swelling values.

Further experimental evidence was observed in two fuel pins clad with Type 316 stainless steel and loaded with fuel of different densities (83.9% T.D. powder fuel and 86.7% T.D. pelleted fuel). The pin with the higher density fuel was found to have measurable cladding creep strain as compared to no strain for the pin containing low density fuel. The higher density fuel with its higher effective swelling rate had caused a higher stress in the clad. It was found that the cladding from this pin had swollen more than anticipated from unstressed data suggesting that some stress enhancement of swelling occurred in the higher stressed cladding [36].

Theoretical studies of the effects of stress on voids within grains, based on the work of Harrison [37] and Gulden and Kaae [8], indicate that under high fast flux irradiation ($\sim 10^{15}$ n/cm²/sec with $E > 0.1$ MeV) void growth can occur at $\sim 0.5T_m$ due to stress motivated vacancy flow to voids. Under conditions of hydrostatic tension the stress gradients developed around voids provide an extra driving force for void growth. Irradiation experiments are planned in EBR-II to investigate the theoretical predictions.

In-Reactor Creep

Theoretical - A computer program has been written which uses the Li-Harkness model to calculate the void volume and average void and loop sizes. This program emphasizes the need to separate neutron flux and irradiation time in describing void swelling. It is also useful as a swelling or creep subroutine in fuel element model calculations and to evaluate the effects of microstructural variables such as carbide precipitation which may reduce void formation at temperatures near $0.5T_m$.

The point defect - dislocation interactions proposed in the model will enhance climb or Weertman-type creep and results in the following equation for irradiation induced creep [17]:

$$\dot{\epsilon} = \alpha \sigma^2 L / \mu^2 d \left[D_s / b e^{-\sigma^2 L b^2 / \mu k T} + K J_{xs} \right] \quad (3)$$

This equation at higher fluences predicts that voids and dislocation loops will build up in concentration and present additional barriers to dislocation motion, tending to decrease the creep rate as shown in Figure 7.

Experimental - Experimental irradiation induced creep observations have been reported in several types of austenitic stainless steel irradiated in several thermal and fast reactors [18-24]. Since the discovery of metal swelling and the design related problems associated with this phenomena such as bowing of in-core components, it has been necessary to estimate the magnitude of irradiation induced creep and stress relaxation which might be expected to occur in a fast reactor spectrum. Such an analysis has been completed using existing data from several investigators, including Gilbert [35] and Mosedale [19], and have resulted in the following equations (4-5) representing a statistical fit of data on solution treated and cold worked austenitic stainless steel, respectively. These data were adjusted for spectral difference by procedures developed by McElroy and co-workers [38] and indicate that the cold worked material can be expected to creep more than the solution treated material for any given fluence level and that irradiation induced creep is a linear function of stress.

$$\bar{\epsilon} / \bar{\sigma} = 0.2 \times 10^{-8} + 1.87 \times 10^{-30} \alpha \phi t \quad (4)$$

$$\bar{\epsilon} / \bar{\sigma} = 0.8 \times 10^{-8} + 1.86 \times 10^{-30} \alpha \phi t \quad (5)$$

An instrumented creep capsule has been fabricated for operation in the EBR-II. This capsule, to be located in Row 5 position in the EBR-II simultaneously measures stress and strain as a function of time on a uniaxial specimen of 20% cold worked Type 316 stainless steel at a controlled temperature. Four biaxial stress rupture specimens held at four controlled stress levels are also within the test chamber and postirradiation examination will allow determination of average in-reactor creep rates which occurred during the irradiation.

Stainless steel tubes sealed under a predetermined pressure are being irradiated in the EBR-II in contact with the reactor sodium coolant [22]. After each reactor cycle the tubes are unloaded from the reactor, carefully measured and replaced in the reactor. One test currently in the reactor achieved a maximum fast fluence of 9.9×10^{21} n/cm² (E > 0.1 MeV). It contains 18 annealed Type 304L tubes, 7.25 mm OD, 0.5 mm wall thickness and 150 cm long. Hoop stresses are in the range 10 to 30 kpsi at the 370°C sodium inlet temperature. The tubes extend 75 cm above and below the core centerline so that the fast flux varies along the length from about 1.0×10^{15} to 1.0×10^{13} n/cm²/sec. Profile measurements at a peak fluence of 3×10^{21} n/cm² show that there is a considerable orientation dependence of the measurements due to non-uniformity arising from the tube fabrication process. This and the contribution from void swelling must be considered in assessing the

creep strain; void swelling is derived from measurements on unstressed tubes. The temperature profile along the tubes is linear. Hence, it is possible to compare the creep rate in the low flux parts of the tubes with the rate in the high flux regions and derive a value for radiation enhanced creep. At 30,000 psi an average creep rate of $1 \times 10^{-6} \text{ hr}^{-1}$ was calculated from the model described above (equation 4) so that the experimental and theoretical values are in fair agreement.

IV. STRUCTURAL ANALYSIS AND CORRELATION

The metallurgical structure of austenitic stainless steels is known to change as a result of exposure to fast neutrons, temperature and sodium environments. Brager, et al [12] using transmission electron microscopy have completed a description of the defects produced in solution treated and cold worked Types 304 and 316 stainless steel irradiated in the EBR-II over a 370-700°C temperature range to a maximum fluence of about $6 \times 10^{22} \text{ n/cm}^2$ ($E > 0.1 \text{ MeV}$). In general the investigators have observed the following.

Void Size - Voids have been found in solution treated Types 304 and 316 stainless steels which were irradiated over the temperature range 370°C to 650-700°C (0.38 Tm to 0.58Tm) [39]. In general, the minimum, mean, and maximum observed void diameters increase with increasing temperature but increase only slightly with increasing neutron fluence over the range of 0.5 to $5 \times 10^{22} \text{ n/cm}^2$ ($E > 0.1 \text{ MeV}$). Smaller void sizes, as well as smaller void number densities and less swelling, are found in solution treated Type 316 stainless steel than in Type 304 stainless steel.

Void Number Density - The flux rate effect on void formation, particularly over the range of fluxes found in fast reactors, appears to be small [12,40]. The void number density in solution treated Type 304 stainless steel follows a power law relationship of fluence with the power increasing with irradiation temperature and the mean value being near unity [41]. Data on samples irradiated at high temperatures (550 to 650°C) indicate that void nucleation ceases after low void number densities have been formed [27,42].

Effects of Cold Work - The void number density in 27% cold worked Type 316 stainless steel is about 1/50 of that in the solution treated steel. However, the mean void diameters of the cold worked and solution treated steels are nearly the same. Frank faulted loops are produced in the cold worked structure over the same temperature range as in the solution treated steels. The 27% cold worked Type 316 stainless steel did not recrystallize at temperatures of 850°C although the dislocation density was reduced for irradiation temperatures above 650°C [43].

Loop Distribution - Frank faulted loop sizes increase with increasing irradiation temperatures but do not appear to change significantly with increasing fluence. At low irradiation temperatures (370 to 450°C) faulted loops are formed with sizes that increase and number densities

that decrease with increasing temperature. From 450 to 600°C faulted loops co-exist with perfect loops and with a dislocation network formed by interaction of unfaulted loops. Above 600°C only a dislocation network exists.

Precipitates - Coherent cuboidal $M_{23}C_6$ and ribbon shaped sigma phase precipitates are formed uniformly throughout the matrix of the solution treated Types 304 and 316 stainless steel. At high irradiation temperatures (700 to 900°C where neither voids nor sigma phase precipitates are present) $M_{23}C_6$ is nearly continuously precipitated along grain boundaries.

As a final point, recent data obtained in current sodium coolant technology programs show degradation of stress rupture life (50% reduction) and ductility as a result of severe losses of carbon, nitrogen and boron from Types 304 and 316 stainless steel when exposed to flowing sodium under stress at 704°C. Loss of carbon is accompanied by the formation of sigma phase particles at the grain boundaries of the specimens, believed to be responsible for the degradation of mechanical properties observed [44].

V. POSITIVE ION BOMBARDMENT OF AUSTENITIC STAINLESS STEEL

Several major uncertainties exist regarding metal swelling and irradiation induced creep including: (1) the nucleation and growth kinetics, (2) swelling at fluences exceeding 1×10^{23} n/cm², (3) effects of material composition and metallurgical structural, (4) saturation effects and (5) kinetics associated with irradiation induced creep. To evaluate these uncertainties bombardment by positive ions has been used to simulate neutron irradiation.

Proton Bombardment - Before irradiation specimens were uniformly injected with helium to a concentration of about 5 atomic parts per million by means of a cyclotron [45] to simulate reactor conditions during the proton bombardment at energies of either 0.75 or 1.0 MeV. The proton fluences were varied between 6×10^{17} and 1.3×10^{20} P/cm² at a flux of about 1.5×10^{15} P/cm²-sec. The number of displacements for a given proton fluence and depth of proton penetration was calculated from the Kinchin-Pease model with a displacement threshold energy of 25 eV [46]. The proton energy as a function of penetration into the sample was determined from the work of Janni [47].

The increase in sample volume per unit volume (swelling) as a function of calculated number of atom displacements is shown in Figure 8. The horizontal bars through the data points represent the variation in calculated atom displacements resulting from ± 1000 Å uncertainty in foil position along the proton path. The vertical bars represent the uncertainties encountered in determining the swelling from several photomicrographs of the same foil. These uncertainties are frequently well beyond the subjective error expected in operation of the particle size analyzer and are thought to be real. Such variations in swelling may result from slight microstructural inhomogeneities within small regions of the sample.

The line through the data points in Figure 8 was drawn according to a least squares fit per atom and is represented by equation (7).

$$\Delta V/V (\%) = 0.17 N^{1.2} \quad (7)$$

where $\Delta V/V$ is the swelling, and N is the atom fraction of displacements. The swelling dependence given by equation (7) is in reasonable agreement with a theory for void growth recently proposed by Wiedersich [10]. It is assumed that void nucleation occurs early in the irradiation and that the principal dislocation sinks are loops which nucleate and grow by precipitation of excess interstitials during irradiation. It was previously determined that the loops present in these samples are interstitial in character [48].

The effect of displacement rate on observed void density is shown in Figure 9 and appears to be independent of fluence. These data are for irradiations where there were different total numbers of displacements/atom. No dependence of void density (number-cm⁻³) on the number of displacements/atom has been observed. It appears from the results in Figure 9 that higher displacement rates enhance the nucleation of voids, presumably early in the irradiation.

That void nucleation does occur at early irradiation times is further evidenced by the observation that in the present work the minimum observed void diameter increases with increasing number of displacements/atom. For example, at about 1 displacement per atom the minimum void size is about 40 Å, while it is about 95 Å at 50 displacements per atom. One specimen containing no pre-injected helium has been irradiated at 500°C. In a region which underwent about 5 displacements per atom, an extremely low density of fairly large voids was present. Helium appears to have a significant effect on the stability of void nuclei.

Heavy Ion Bombardment - Solution annealed and 20% cold work Type 316 stainless steel specimens have been bombarded with 5 MeV Ni⁺⁺ ions. The nature of the defects produced by ion bombardment is the same as in the case of neutron irradiation, but it is not yet clear whether a direct correlation can be made between the two cases. The dislocation structure observed to form at the various temperatures indicates a high density of loops ($\sim 10^{14} - 10^{15}$ cm⁻³) but a lack of faulted loops at 475°C which are known to exist in neutron irradiated material. Also, it appears that few, if any, voids are formed at temperatures as low as 475°C. Voids with an average size of 300 Å and density of 3.3×10^{13} cm⁻³ were observed after bombardment at 585°C.

When annealed Type 316 stainless steel was bombarded with 5 MeV Ni⁺⁺ at a constant temperature of $\sim 595^\circ\text{C}$ at fluences of 3.6×10^{15} , 1.2×10^{16} and 3.4×10^{16} ion/cm² faulted dislocation loops were formed. The dislocations appear to be aligned preferentially along carbide precipitates and grain boundary substructure along {111} crystallographic planes. The dislocation loop density increased from 0.8×10^{14} cm⁻³ to 1.5×10^{14} cm⁻³ as a result of the increased

FIGURE 5. EFFECT OF IRRADIATION ON THE HIGH TEMPERATURE BIAxIAL DUCTILITY OF TYPE 304 TUBING

FIGURE 7. THE EXPECTED CHANGE IN THE 400°C IN-REACTOR CREEP RATE OF TYPE 304 STAINLESS STEEL WITH FLUENCE

FIGURE 9. VOID DENSITY AS A FUNCTION OF DISPLACEMENT RATE FOR PROTON-IRRADIATED 316 STAINLESS STEEL CONTAINING HELIUM

FIGURE 6. THE TEMPERATURE DEPENDENCE OF VOID SIZE AND NUMBER DENSITY IN SOLUTION ANNEALED TYPE 304 STAINLESS STEEL IRRADIATED IN EBR-11

FIGURE 8. SWELLING AT 500°C AS A FUNCTION OF DAMAGE FOR PROTON-IRRADIATED 316 STAINLESS STEEL CONTAINING HELIUM

fluence. The average size was 300 Å, 600 Å and 350 Å for the lowest, intermediate and highest fluence, respectively. The exact reason for this reduced loop size at the highest fluence is unknown. It may possibly be related to the interaction of the loops at the higher dislocation density and the formation of voids observed at this fluence and temperature region.

Effects on Irradiation Induced Creep - Harkness and Yaggee are conducting experiments towards evaluation of irradiation induced creep by means of ion bombardment. The ion beam is a 22 MeV deuteron beam, which at 4 μA, creates damage at about the same rate as a flux of 2×10^{15} n/cm²/sec in the EBR-II reactor. The beam is modulated by a rotating metal 'wedge' so that damage is created fairly uniformly through the test sample. The results to date indicate that creep enhancement does occur under deuteron bombardment. At about 420°C and 50,000 psi stress, the creep rate is enhanced about one order of magnitude. The minimum creep rate ($\dot{\epsilon}_m$) increases from $< 1 \times 10^{-6}$ hr⁻¹ without deuteron bombardment to about 2×10^{-5} hr⁻¹ with deuteron bombardment. A uniaxial creep test has been performed at 370°C during 22 MeV deuteron bombardment on a sample of Type 304 stainless steel previously irradiated in EBR-II to a total dose of 8×10^{21} n/cm² at 460°C. The purpose of the test was to determine the effect of fluence on the in-reactor creep properties. The results showed irradiation in EBR-II reduced the creep rate from 2.5×10^{-4} to 1.1×10^{-5} hr⁻¹.

VI. CONCLUSIONS

The design of fast breeder reactors presents unique problems associated with materials behavior. These problems include the synergistic effects of sodium environment at elevated temperature, irradiation effects on mechanical properties and metal swelling. The data to date indicate that a reduction in both stress rupture and ductility properties of austenitic stainless steel may be expected to occur at end of life for in-core components. In addition, allowance for metal swelling and irradiation induced creep must be made for components exposed to high fluence levels. As a result of the on-going programs mechanical property data is available to provide guidance in establishing material limits and design criteria and interim design equations describing the fluence and temperature dependency of metal swelling and irradiation induced creep are available. As a result of the overall National effort materials technology of fundamental and engineering significance will make possible the design, construction and operation of reliable LMFBR systems to economical burnup levels.

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- Figure 1: The Effect of Irradiation on the Yield Stress of Types 304 and 316 Stainless Steel When Tested at the Irradiation Temperature.
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- Figure 9: Void Density as a Function of Displacement Rate for Proton-Irradiated Type 316 Stainless Steel Containing Helium.

REFERENCES

- [1] CARLANDER, R., HARKNESS, S. D., YAGGEE, R. L., Nuclear Applications and Technology, 2, (July 1969), 67-75.
- [2] LAURITZEN, T., WITHOP, A. W., FERGUSON, G. P., "Mechanical Properties Evaluation of Austenitic Stainless Steels Irradiated in EBR-II", GEAP-10066, (July 1969).
- [3] LOVELL, A. J., HOLMES, J. J., Personal Communications, (April, 1971)
- [4] BLOOM, E. E., STIEGLER, J. O., "Effect of Fast Neutron Irradiation on the Creep Poture Properties of Type 304 Stainless Steel at 600°C", ORNL-114-3169, (January, 1971), Presented at the 73rd Annual ASTM Meeting, June 1970 Niagara Falls, Canada.
- [5] LOVELL, A. J., BLACKBURN, L. D., BARKER, R. W., Submission for WADCO Quarterly Technical Report, January, February, March (1971).
- [6] HARKNESS, S. D., LI, CHE-YU, "A Study of Void Formation in Fast Neutron Irradiated Metals - To be published in Metallurgical Transactions.
- [7] CLAUDSON, T. T., HOLMES, J. J., STRAALSUND, J. L., BRAGER, H. R., "Fast Reactor Radiation Induced Changes in Cladding and Structural Materials", Radiation Damage in Reactor Materials, IAEA, Vol. II, Vienna, (June 1969), 165-187.
- [8] Gulden, T. D., KAAE, J. L., Journal Nuc. Mat. 32, (1969) 168.
- [9] SULLOUGH, R., EYRE, B. L., PERRIN, R. G., Nuc. App. & Tech. Vol. 9, No. 3, (1970).
- [10] WIEDERSICH, H. W., "On Void Formation During Irradiation", Second International Conference on the Strength of Metals and Alloys, Asilomar, Cal. (Sept. 1970).
- [11] HAM, F. S., J. Appl. Physics, Vol. 30 No. 6, (June 1959), 915-926.
- [12] BRAGER, H. R., STRAALSUND, J. L., HOLMES, J. J., BATES, J. F., "Irradiation Produced Defects in Austenitic Stainless Steel", WHAN-FR-16, To be published.
- [13] BECKER, R., DORING, W., Ann. Phys. 24, (1935) 719.
- [14] HARKNESS, S. D., LI, C. Y., "Radiation Damage in Reactor Materials" Vol. II, IAEA Vienna, (1969), 169.
- [15] BLEWITT, T. H., Private Communications.
- [16] ANSELL, G. D., WEERTMAN, J., Trans. AIME, Vol. 215, (1959) 838.
- [17] HARKNESS, S. D., TESK, J. A., LI, C. Y., J. Nuc. Appl. & Tech. No. 9, (July 1970), 24.
- [18] HESKETH, R. V., "Proceedings of the International Conference on Solid-State Physics Research with Accelerators", BNL-50083, Brookhaven National Laboratory, Upton, N.Y., (1967).
- [19] MOSEDALE, D., LEWTHWAITE, G. W., LEET, G. O., SLOSS, W., Nature, Vol. 224, (1969), 1301.
- [20] LEWTHWAITE, G. W., MOSEDALE, D., WARD, I. R., Nature, Vol. 216 (1967), 472.
- [21] GILBERT, E. R., Unpublished data, WADCO Corporation, Richland, Washington.
- [22] WALTERS, L. D., WALTER, C. M. PUGACZ, M. A., TESK, J. A., CARLANDER, R., LI, C. Y., Trans. Am. Nucl. Soc., Vol. 13, (1970), 145.
- [23] JOSEPH, J. W., "Stress Relaxation in Stainless Steel During Irradiation", DP-3c9, E. I. DuPont de Nemours & Co., Aiken, S. Carolina, (1959).
- [24] SCHREIBER, R. E., "Relaxation of Torsional Stresses in Stainless Steel During Irradiation", DP-669, E. I. DuPont de Nemours & Co., Aiken, S. Carolina, (1962).
- [25] STRAALSUND, J. L., BATES, J. F., BIERLEIN, T. K., Personal Communications (April 1970).
- [26] APPLEBY, W. K., SANDUSKY, D. W., WOLFE, V. E., "Fluence and Temperature Dependence of Void Formation in Highly Irradiated Stainless Steels", Conference on Radiation Induced Voids in Metals, Albany, New York, (June, 1971).
- [27] SANDUSKY, D. W., WOLFE, V. E., SPALARIS, C. N., "Irradiation Temperature Dependence of Void Formation in Type 304 Stainless Steel", submitted to J. Nucl. Matl.
- [28] GARNER, F., BOLTAX, A., HARBOURNE, B. L., BIANCHERIA, A., "The Effect of Stress on Radiation Induced Void Growth", To be published, Presented at the International Conference on Radiation Induced Voids in Metals, (June, 1971).
- [29] HYAM, E. D., SUMNER, G., IAEA Conference on Radiation Damage in Solids, Vol. 1, (1962), 323.
- [30] ROWCLIFFE, A. F., CARPENTER, G. J. C., MERPICK, H. F., NICHOLSON, R. B., ASTM Spec. Tech. Publ. (1967), 426.
- [31] STIEGLER, J. O., FARRELL, K., MCCOY, H. E., Jnl. Nuc. Mat. 25, (1968), 340.
- [32] GARR, K. R., KRAMER, D., PHODES, C. G., AIME Metallurgical Trans. Vol. 2, (January, 1971), 260.
- [33] YAGGEE, F. L., STYLES, J. W., "Irradiation Effects on Reactor Structural Materials", Quarterly Progress Report - August-September-October, (1970), 3.21.
- [34] BOLTAX, A., BIANCHERIA, A., HARBOURNE, B. L., SOFFA, T. P., "Mixed Oxide Fuel Pin Performance Analysis Using the OLYMPUS Code", To be published, Presented at the AYS Conference on Fast Reactor Fuel Element Technology, (April, 1971).
- [35] GILBERT, E. R., BLACKBURN, L. D., "Irradiation Induced Creep in Austenitic Stainless Steel", WHAN-FR-30, (October, 1970).
- [36] Sodium Cooled Reactors Fast Ceramic Reactor Development Program- 37th Quarterly Report, November 70 January 71, GEAP 10028-37 (March, 1971), 71.
- [37] HARRISON, J. W., Jnl. Nucl. Mat. 23, (1967), 139.
- [38] MCELROY, W. N., DAHL, R. E., GILBERT, E. R., Nuclear Engineering and Design, (1970).
- [39] BRAGER, H. R., STRAALSUND, J. L., "The Microstructure of Fast Reactor Irradiated Type 316 Stainless Steel", J. Metals, Vol. 23, (1971).
- [40] BLOOM, E. E., "An Investigation of Fast Neutron Radiation Damage in an Austenitic Stainless Steel", ORNL-4580, (June, 1970)
- [41] BLOOM, E. E., STIEGLER, J. O., "Recent Observations of Irradiation Induced Void Formation in Austenitic Stainless", Trans. AYS, Vol. 12, No. 2, (December, 1969), 589-590.
- [42] STIEGLER, J. O., BLOOM E. E., "Effects of Thermal Mechanical Treatments on Void Formation in Irradiated Stainless Steel", submitted to J. Nucl. Matl.
- [43] BIERLEIN, T. K., "Damage Structural Characterization and Analysis", WADCO Technical Quarterly Report - October, November, December, (1970).
- [44] SPALARIS, C. N., ZEBOSKI, E. L., "Considerations of Stabilized Stainless Steel Alloys for LMFBR Fuel Cladding", TRANS. AYS, Supplement No. 1, Vol. 14, New Orleans, La. (April, 1971)
- [45] KRAMER, D., BRAGER, H. R., PHODES, C. G., PARD, A. G., J. Nucl. Mat. 25, (1968), 121.
- [46] KINCHIN, G. H., PEASE, R. S., Rep. on Prog. in Phys., 18, (1955), 1.
- [47] JAHNI, J. F., AFWL-TR-65-150 (1960).
- [48] KEEFEP, D. W., ET AL, "Irradiation Effects on Structural Alloys for Nuclear Applications", ASTM-STP 484, ASTM, Philadelphia, (1971).