Proton Irradiation Creep of FM steel T91

Cheng Xu, Gary S. Was

University of Michigan, 2355 Bonisteel Blvd, Ann Arbor, MI 48109-2104, USA

Abstract

Ferritic-martensitic (FM) steel T91 was subjected to irradiation with 3MeV protons while under load at stresses of 100MPa-200MPa, temperatures between 400°C-500°C, and dose rates between 1.4×10^{-6} dpa/s- 5×10^{-6} dpa/s to a total dose of less than 1dpa. Creep behavior was analyzed for parametric dependencies. The temperature dependence was found to be negligible between 400°C – 500°C, and the dose rate dependence was observed to be linear. Creep rate was proportional to stress at low stress values and varied with stress to the power 14 above 160 MPa. The large stress exponent of the proton irradiation creep experiments under high stress suggested that dislocation glide was driving both thermal and irradiation creep. Microstructure observations of anisotropic dislocation loops also contributed to the total creep strain. After subtracting the power law creep and anisotropic dislocation loop contributions, the remaining creep strain was accounted for by dislocation climb enabled by stress induced preferential absorption (SIPA) and preferential dislocation glide (PAG).

Keywords:

Creep, irradiation, ferritic martensitic steels, T91, transient creep.

Corresponding author: Cheng Xu
University of Michigan
2355 Bonisteel Blvd, Ann Arbor
MI, 48109-2104

Phone: 7346213647
Email: chengxu@umich.edu

1. Introduction

Ferritic-martensitic (F-M) steels have a number of attractive properties for use as cladding and ducts in sodium cooled fast reactors, including high strength at elevated temperatures, good compatibility with sodium coolant, and excellent swelling resistance under irradiation. At fast reactor operating temperatures, irradiation creep becomes a major concern. An understanding of the creep mechanism was required to determine the suitability of these steels for the intended application. A large body of work had been done to summarize the potential irradiation creep mechanisms that could occur in FM steels. [1]–[3] The majority of the recent efforts had been focused on the irradiation creep mechanisms of stress-induced preferential absorption (SIPA), [4]–[6], preferential absorption glide (PAG), [7], [8] and preferential emission (PE) at higher temperatures [8]. To determine the dominant irradiation creep mechanism in FM steels, more irradiation data were needed to clarify the dependence of creep on the various irradiation parameters and to link the irradiation creep microstructure to the measured creep strain for a detailed analysis.

Irradiation creep experiments in test reactors such as FFTF, EBR-II, and HFIR [1–3] used pressurized tubes that were periodically removed to measure the dimensional changes as a function of time. These experiments provided valuable quantitative creep data under conditions similar to those during actual operation. However, neutron irradiations were too expensive and time consuming to fully explore the nuances of specific creep mechanisms. In addition, neutron irradiation creep experiments were not conducive to exploring the transient behavior of irradiation creep that act as a precursor

to steady state behavior. Therefore, efforts to systematically narrow down the specific mechanisms of irradiation creep are still needed.

To provide additional insight into the mechanism of irradiation creep of FM steels, experiments were conducted using 3 MeV protons generated by a linear accelerator. Creep behavior at low dose can be captured in real time at damage rates comparable to those of fast neutron irradiations. The instantaneous strain rates measured at low dose were analyzed to systematically explore the effect of single variables, such as dose rate, temperature, and stress. The measured irradiation creep rate parametric dependencies were analyzed in relation to existing irradiation creep theories. The low activation from proton irradiation also made post irradiation analysis easy to perform, providing a unique opportunity to capture microstructure during its evolution

This study explored the parameter dependencies of irradiation creep for FM steel T91 to obtain data that was used to differentiate between possible irradiation creep mechanisms such as preferential emission (PE), stress induced preferential absorption (SIPA), and preferential absorption glide (PAG). Proton irradiation creep experiments were conducted in the Michigan Ion Beam Laboratory (MIBL), and the irradiated samples were analyzed at the Electron Microbeam Analysis Laboratory (EMAL) at the University of Michigan, and the Nanotech User Facility (NTUF), at University of Washington. The results of the dose rate, temperature, and stress dependencies of irradiation creep are discussed along with its implications for existing irradiation creep theories.

2. Experiment

The composition of the T91 heat used in this experiment is given in Table 1. The as-received T91 plate was normalized at 1040°C for 46 minutes, followed by air cooling, then tempering at 760°C for 42 minutes. The samples were made by electric discharge machining into a dog-bone shape with a thickness of 100 μm , as shown in Figure 1. The samples were electro-polished in a 10% perchloric acid, and 90% methanol solution at -40°C. An applied voltage of 20V produced a current density of 2A/cm². The average removal rate of this electro-polishing method was 20 $\mu\text{m}/\text{min}$, and the samples were polished to a final thickness of 35 $\pm$ 2 μm to ensure full penetration of the 3MeV protons. The damage profile of SRIM simulation is illustrated in Figure 2. The damage profile was fairly flat at an average of 5.6 $\times 10^{-5}$ displacements/angstrom-ion, with the exception of last 2 μm in the back where the damage was factor of 5 higher than the rest of the sample thickness. Because the higher damage was limited to a thin region close to the surface where radiation damage was unlikely to accumulate, the difference in damage was not expected to adversely affect the bulk irradiation creep. For the purpose of this study, the dose rate was calculated using the average damage profile across the sample thickness.

Irradiation creep experiments were conducted in a specially made irradiation creep stage described in a previous paper by Xu and Was [9]. Irradiations were conducted using 3 MeV protons, which have a penetration distance of 40 μm in stainless steel, to ensure that there was no implantation within the 35 μm sample. The experimental setup, shown in Figure 3, had the capability for in-situ strain measurements for a dog-bone sample under uniaxial tensile stress, high temperature, and proton irradiation. The strain rate measured by each irradiation creep experiment was obtained by taking the

linear fit of the strain over time as measured by a laser speckle extensometer in the gage length. The irradiation temperature were monitored by a 2D thermal imager, and applied stress monitored by a load cell.

Seventeen irradiation creep experiments in total were conducted between 400°C to 500°C, at dose rates between 1.4×10^{-6} and 5×10^{-6} dpa/s (as determined by SRIM Kinchin-Pease Mode [10]) to doses of <1 dpa and under loads of 100 – 200 MPa. The strain rate data were analyzed as a function of dose rate, temperature, and stress. Error in the irradiation creep strain rates was determined by repeated experiments under the same conditions and on two samples, designated A and B. The thermal creep test condition of 500°C at 200MPa (TT500200-A and TT500200-B) was repeated, and two irradiation creep test conditions, 450°C at 200MPa (IT450200-A and IT450200-B) and 450°C at 180MPa (IT450180-A and IT450180-B) were also repeated. The calculated standard errors were found to vary between 29.4% to 50.5% of the measured creep rates. The largest percentage error was chosen as a conservative estimate of the error from repeated experiments for all measured creep rates.

Microstructure analysis of the irradiation creep samples was conducted using JOEL 3100 and Tecnai G2 transmission electron microscopes (TEM) to obtain information on the orientation of dislocation loops. The TEM samples were made using focused ion beam (FIB) milling to lift out a sample that preserved the tensile axis orientation as shown in Figure 1. Samples were imaged in the <001> zone axis under a two-beam condition with $g = \langle 110 \rangle$ such that two sets of edge-on $a_0\langle 100 \rangle$ loops were visible. The angle θ between the normal vector of the dislocation loop plane and the tensile axis was used to define the loop orientation as shown in Figure 4. The number of

dislocation loops of the two sets of edge-on loops were measured as a function of the loop orientation to the tensile axis to quantify its anisotropy.

3. Results

Seventeen irradiation creep experiments were conducted for this study at various temperature, stress, and dose rate conditions. The typical irradiation creep strain over time data is shown in Figure 5. The sample temperature, applied load, dose rate, and sample strain were all continuously monitored throughout the irradiation. Strain rates were calculated from line fits to the strain over time where temperature, dose rate, and stress were kept constant. The strain rates measured for each condition are tabulated in Table 2. The strain rates measured were then analyzed as a function of their dose rate, temperature, and stress dependence.

3.1 Dose Rate Dependence

An experiment was conducted on sample IT500160 to explore the dose rate dependence at 500°C and 160MPa in real time, taking advantage of the in-situ strain measurement capability of the facility. A high irradiation temperature was necessary to accommodate for the large range of beam heating required for the different dose rates. Three dose rate conditions were used in this experiment: 1.5×10^{-6} dpa/s, 1.7×10^{-6} dpa/s, and 2.4×10^{-6} dpa/s. The corresponding strain rates were found to be $1.78 \pm 0.89 \times 10^{-9} \text{ s}^{-1}$, $2.33 \pm 1.67 \times 10^{-9} \text{ s}^{-1}$, and $5.08 \pm 2.54 \times 10^{-9} \text{ s}^{-1}$. The strain rates are plotted against the dose rates in Figure 6. The data point at a dose rate of zero is the measured thermal creep rate at that temperature and the fit includes the data through these four points. A linear fit to

the dose rate data fall within the experimental errors, suggesting the possibility of a linear dose rate dependence . The implication of a linear dose rate dependence and alternative dose rate dependencies are addressed in detail in the discussion section.

3.2 Temperature Dependence

Irradiation creep experiments were conducted at 400°C, 450°C, and 500°C at 160MPa to explore the temperature dependence. The dose rate was maintained at the nominal value of 1.7×10^{-6} dpa/s with the exception of the 400°C experiment where a lower dose rate of 1.3×10^{-6} dpa/s was needed to maintain the temperature. The strain rates measured for the experiments at 400°C, 450°C, and 500°C were $2.9 \pm 1.45 \times 10^{-9} \text{ s}^{-1}$, $2.7 \pm 1.35 \times 10^{-9} \text{ s}^{-1}$, and $2.33 \pm 1.17 \times 10^{-9} \text{ s}^{-1}$ respectively. Because the creep rate measured at 400°C was conducted at a slightly lower dose rate, extrapolation of the 400°C strain rate to 1.7×10^{-6} dpa/s was calculated using the dose rate dependence at 500°C. The resulting strain rate was $2.93 \times 10^{-9} \text{ s}^{-1}$. The strain rates are plotted as a function of temperature shown in Figure 7. The proton irradiation creep rates exhibited no temperature dependence.

3.3 Stress Dependence

To determine the stress dependence of irradiation creep, seven experiments were conducted at a constant temperature of 450°C and a dose rate of 1.7×10^{-6} dpa/s. The applied stress ranged from 100MPa to 200MPa at 20MPa intervals. The stress dependence of irradiation creep is shown in Figure 8. The creep rate dependence on stress

appeared linear below 160MPa; but above that value, the creep rate increased rapidly with stress, indicating a high stress exponent.

Determination of the stress exponent in both stress regimes was done recursively by minimizing the residuals of the curve fit to the data, assuming the following form of the creep rate dependence:

$$\dot{\epsilon} = B_I \sigma^{n_I} + B_T \sigma^{n_T} \quad (1)$$

where B_I is an effective irradiation creep compliance, σ is the applied stress, and n_I is the irradiation creep stress exponent. Similarly, B_T is the effective thermal creep compliance, σ is the applied stress, and n_T is the thermal creep stress exponent. The best fit for the data yield the following constants: $n_I=0.86$, $B_I=3.2 \times 10^{-11} \text{s}^{-1} \text{MPa}^{-0.86}$, $n_T=14$, and $B_T=5.4 \times 10^{-41} \text{s}^{-1} \text{MPa}^{-14}$. Separately, the irradiation and thermal creep describe the asymptotic behavior of the data, with irradiation creep at low stress and thermal creep at high stress. By taking the summation of both fits, the total irradiation creep rates are described by the following equation:

$$\dot{\epsilon} = 3.2 \times 10^{-11} \sigma^{0.86} + 5.4 \times 10^{-41} \sigma^{14}. \quad (2)$$

3.4 Microstructure

The loop microstructures of four irradiation creep experiments were analyzed in TEM. The major effect of irradiation was the formation of dislocation loops for which the number density varied with orientation. The loops were predominantly of the $a_0 \langle 100 \rangle$ type. Figure 9 shows representative TEM images from the four irradiation creep samples. In order to make appropriate comparisons between the loop densities from different grains, the loop number density for each orientation of $a_0 \langle 100 \rangle$ loops was normalized by

the total number of $a_0 \langle 100 \rangle$ loops in that grain. The anisotropy in the loop density was defined by the fraction of one set of $a_0 \langle 100 \rangle$ loops over the entire $a_0 \langle 100 \rangle$ loop population of that grain. This normalized dislocation loop density is plotted against angle θ for all samples in Figure 10. Due to the small loop size of the $a_0/2 \langle 111 \rangle$ loops, they were not analyzed in this study. The measurements showed that there was strong anisotropy in the $a_0 \langle 100 \rangle$ dislocation loop density, in favor of loops that add extra half planes in the direction of the tensile stress. There was no anisotropy in the loop size, and no voids were observed in any of the samples.

The strain due to the loop anisotropy for each sample was calculated using an equation developed by Kroupa [11] assuming a continuous distribution of interstitial loops. The result of the calculations are summarized in Table 3. The strain contribution from loop anisotropy in the tensile direction was found to be responsible for 4-11% of the total measured creep strain. Details of the calculations were described in a previous paper by Xu and Was [12].

4. Discussion

Several key observations were made as the result of this study. Dose rate dependence of irradiation creep was found to be linear, temperature dependence of irradiation creep was found to be negligible between 400°C - 500°C, and stress dependence of irradiation creep at 450°C was best described by an approximately linear stress dependence at low stress and a large stress exponent at high stress. Irradiation creep microstructure consisted of anisotropic distribution of dislocation loops. These

results are discussed in comparison to previous irradiation creep observations for FM steels and irradiation creep mechanisms.

4.1 Dose Rate Dependence

The linear dose rate dependence observed for proton irradiation creep is generally expected in irradiation creep of FM steels. Chen [13], [14] normalized the strain rates of ferritic ODS alloys irradiated by light ions from a compact cyclotron to obtain creep compliance by assuming a linear dose rate dependence. Chin [15] also assumed a linear dose rate dependence for HT9 irradiated in EBR-II. Although there are no neutron irradiation creep experiments specifically targeting the dose rate dependence of FM steels, Grossbeck [16] conducted irradiation creep experiments for three cold-worked austenitic steels irradiated in different mixed spectrum reactors for various dose rates. The slope of the fits on a log scale was approximately 0.5 in all cases, suggesting a square root dose rate dependence for neutron irradiated austenitic steels.

The difference between the dose rate dependence of austenitic steels and FM steels is due to the difference in their steady state point defect concentrations. Depending on the total sink density, the point defect concentrations can exhibit either a linear dependence or a square root dependence [17]. When the recombination of behavior of point defects dominates, the steady state defect concentration has a square root dependence on dose rate. In the sink dominant regime, the steady state defect concentration varies linearly with dose rate.

Characterization of the irradiated microstructure for T91 yields a total sink strength of $7.15 \times 10^{15} \text{m}^{-2}$ by accounting for sub-grain boundaries, dislocation loops,

dislocation lines, and incoherent precipitates for sample IT450200, Table 4. The observed high sink densities of FM steels result in a much shorter time constant for interstitials to reach sinks vs. recombining with vacancies. The result is a sink dominant case that is consistent with the roughly linear dose rate dependence. By contrast, the sink density in austenitic stainless steels is expected to be much lower due to the relatively uncomplicated microstructure. Therefore, a square root dependence on dose rate is reasonable for this austenitic alloys.

Proton irradiation creep experiments at various dose rates suggest a linear dose rate dependence for FM steel T91. Analysis of the sink densities based on irradiation creep microstructure provided more evidence in support of that claim. The results of this study showed that dose rate dependence of irradiation creep experiments are effected by the microstructure, and provided supporting evidence for the historical assumption that FM steels exhibit a linear dose rate dependence. [13], [14]

4.2 Temperature Dependence

Temperature dependence of proton irradiation creep was consistent with existing observations from reactor irradiations [1]–[3], [13], [18]. Figure 11 plots the strain rates observed for a set of 9Cr-1Mo pressurized tube experiments conducted by Tolockzo [18] in FFTF along with the proton irradiation creep rates. The reactor irradiation creep strain rates found below 500°C appeared to have a small negative temperature dependence similar to those found in this study. However, the difference in the strain rates were small enough that the temperature dependence of irradiation creep can be considered to be negligible.

Although the temperature dependence of proton irradiation creep rates match very well to those of neutrons, the magnitude of the strain rates were much higher. Under fast reactor conditions, the minimum creep rate for FM steels at 400-433°C for 100MPa was observed to be about $7 \times 10^{-6} \text{ dpa}^{-1}$. Assuming the average dose rate of $1 \times 10^{-6} \text{ dpa/s}$ in reactor, this corresponds to a creep around $7 \times 10^{-12} \text{ s}^{-1}$. [18] Proton irradiations at 450°C and 100MPa at dose rate of $1.7 \times 10^{-6} \text{ dpa/s}$ yielded a creep rate of about $1.67 \times 10^{-9} \text{ s}^{-1}$. This difference persists if the creep rates are normalized by stress and dose rate to get a creep compliance, B . At 500°C, assuming no contribution from irradiation swelling due to the low dose, B_0 for proton irradiation creep is on the order of $1 \times 10^{-5} \text{ MPa}^{-1} \text{ dpa}^{-1}$, while neutron irradiation creep compliance are around $0.5 \times 10^{-6} \text{ MPa}^{-1} \text{ dpa}^{-1}$. [19] This large difference in strain rate is likely due to differences between neutron and proton irradiation creep experiments.

FM steels follow a three stage creep curve with a very short secondary regime. Neutron irradiation creep experiments typically report the minimum creep rate, measured at doses above 100 dpa. In contrast, proton irradiation creep rates were measured at a dose of less than 1 dpa in the primary creep regime. At low dose, the microstructure has yet to develop, and the fewer obstacles against dislocation movement will result in higher strain rates compared to a steady state microstructure. However, the difference in microstructure alone is insufficient to explain the factor of ~100 difference in creep rate between neutron and proton data. The small thickness of the proton irradiation creep samples resulted in a large surface area to volume ratio and could have also contributed to the difference in strain rate. A larger surface area to volume ratio allows dislocations a shorter path to the surface without many prior austenite grain boundaries in the way to act

as obstacles. This could magnify the effect of certain processes that would otherwise be considered negligible, such as thermal creep at below 500°C.

Finally, the difference in damage morphology between neutrons and protons can also add to the difference in the measured creep rate. For a 1MeV neutron, the effective survival rate of the point defects are around 2% while for a 1 MeV proton, it is around 25%. [17] Thus, a higher percentage of point defects survived in proton irradiation experiments than in reactor experiments beyond the differences in dose rates. It has been shown in the dose rate analysis that strain rates are directly related to the point defect concentrations in the material, therefore a higher surviving point defect concentration also contribute to the higher strain rates seen in proton irradiation creep in comparison to neutron irradiations. Further, the larger damage zones created by neutrons vs. protons will contribute to the greater obstacle field in the case of reactor irradiations.

Due to the difference in creep regime, damage morphology, and sample thickness, proton irradiation creep experiments were not directly comparable to neutron data. However, because the differences were consistent in magnitude across all temperature, dose rate, and stress conditions, comparison of parameter dependencies and microstructure features is still valid.

4.3 Stress Dependence

The complex stress dependence found for this study was not typically observed in neutron irradiation creep experiments, where a single stress exponent of magnitude ~ 1.0 was typically used to fit the creep rate [18], [20]. Figure 12 plots the proton irradiation creep rates with the neutron irradiated data using the dose rate dependence in Figure 5 to

extrapolate the damage rates to that of the reactor case. The open symbols represent literature data, and filled data are the results of this study. The dependence on stress over the same stress range is very similar for both irradiations, but the proton irradiation creep rate was about a factor of 100 higher due to the difference in dose and sample thickness discussed earlier.

The high stress exponent observed for proton irradiation creep at stresses above 160MPa was beyond what was measured in neutron irradiation creep experiments. However, a power law exponent of 10 or more was consistently observed in thermal creep of T91 at high stress [21], [22]. This power law breakdown (PLB) behavior has been well documented in the literature [23]. The power law stress dependence suggests that the creep rate is controlled by dislocation glide in the presence of long range internal stress, caused by dislocation networks generating back stress to impede further dislocation glide. Studies on thermal creep behavior show that materials typically enter PLB at stresses above half the yield stress [22], [24]. The yield strength of FM steels at 450°C was found to be around 400MPa [25], therefore PLB should be operating at stresses near 200MPa. The observation of PLB suggested that thermal dislocation creep mechanisms can occur in tandem with irradiation creep at high stress, obscuring the stress dependence of purely irradiation creep mechanisms.

To isolate the irradiation creep strain, the strains predicted by the power law fit (shown as green dashed line in Figure 8) were subtracted from the total strain. To further isolate the creep strains, the contribution from the anisotropic dislocation loop distribution was subtracted from the total measured creep strain. Although no anisotropic loop strain data was available for the 120MPa, 140MPa, and 160MPa conditions,

interpolation of the anisotropic loop strain between the 100MPa (IT450100) and 180MPa (IT450180) conditions was used to estimate those values.

The remaining irradiation creep strains could be due to three irradiation creep mechanisms: preferential emission (PE), stress induced preferential absorption (SIPA), or preferential absorption glide (PAG). Each irradiation creep mechanism has its own unique temperature and stress dependence. The results of the experiments were analyzed to determine the most likely mechanism dominating the irradiation creep.

4.4 Irradiation Creep Mechanism

Preferential emission (PE)

PE is a creep mechanism in which dislocations with Burgers vectors oriented parallel to the tensile axis emit more vacancies than those oriented perpendicular to the tensile axis. [8], [26]. The unequal vacancy emission causes anisotropic dislocation climb that ultimately results in creep strain. Because PE is largely a thermal diffusion mechanism based on the anisotropic emission of vacancies, it becomes rate controlling when thermal defects overtake irradiation defects. Calculations of the defect concentrations for T91 were conducted using point defect balance equations for the sink dominant regime [17]:

$$C_i = \frac{K_o}{K_{is}C_s}, \quad C_v = \frac{K_o}{K_{vs}C_s} \quad (3)$$

where K_o is the dose rate, K_{is} and K_{vs} are the total reaction rates to sinks, and C_s the total sink density. The sink strength include sub-grain boundaries, dislocation networks, dislocation loops, and precipitates. Gupta and Was [27] showed that sub-grains, being the smallest grain feature in FM steels, can greatly affect creep behavior. Therefore, the sub-

grain densities were measured by using line intercept method where interceptions between test lines and sub-grain boundaries were counted to arrive at an average sub-grain diameter. The diameter was then used to calculate a volumetric sub-grain density that serves as the sink strength contribution from sub-grain boundaries. The precipitate measurements were taken from a proton irradiated T91 at 400°C to 1dpa without stress [28]. The precipitates measured from that study were largely incoherent $M_{23}C_6$ carbides on PAG boundaries, therefore they serve as neutral sinks [29]. Capture of defects by dislocations was in the diffusion-controlled regime. Values of sink concentrations and reaction rates are given in Table 4.

The point defect concentrations of T91 as a function of temperature are shown in Figure 13. The calculation shows that thermal vacancies will dominate at temperatures above 540°C, where the temperature dependence will start to increase. This behavior has also been observed in irradiation creep of other FM steels such as HT9 [15], [30] and F82H [31]. Proton irradiation creep experiments on T91 were conducted at and below 500°C, where point defect concentrations are controlled by irradiation. The lack of a dependence of creep on temperature at $T \leq 500^\circ\text{C}$ confirms that PE is not the rate controlling mechanism for proton irradiation creep below 500°C.

Stress Induced Preferential Absorption (SIPA) and Preferential Absorption Glide (PAG)

The dose rate dependence of irradiation creep suggests that it is directly related to the point defect concentrations under irradiation. Analysis on the temperature dependence of irradiation creep eliminated PE, a thermal vacancy diffusion mechanism, as a dominating mechanism. The subtraction of the PLB contribution from the total strain also

removed strain due to vacancy enabled dislocation climb glide. The remaining creep strain is then likely to be due to either SIPA or PAG.

SIPA is a process in which dislocations with Burgers vectors oriented parallel to the tensile axis absorb more interstitials than those perpendicular to it. The anisotropic absorption allows dislocation climb preferentially on planes perpendicular to the tensile axis, thus adding more extra-half planes in the tensile direction to cause strain. Woo [4], [5] proposed that the climb of the existing anisotropic dislocation microstructure would also contribute irradiation creep strain even in the absence of anisotropic diffusion. Mansur [8] expanded on the SIPA mechanism by proposing that dislocations can also glide after preferential climb. SIPA and PAG were described by Mansur [8] as:

$$\dot{\epsilon}_{PAG} = \frac{4}{9} \frac{\epsilon}{b} (\pi L)^{1/2} \Omega D_i C_i \Delta Z_i, \quad (4)$$

$$\dot{\epsilon}_{SIPA} = \frac{2}{9} \Omega L D_i C_i \Delta Z_i. \quad (5)$$

The variable ϵ denotes elastic deflection, which is the applied stress over the modulus σ/E , L is the total dislocation density, Ω is the atomic volume, D_i is the interstitial diffusion constant, and C_i is the steady state interstitial concentration. ΔZ_i is the difference in stress induced anisotropic sink strength for interstitials between aligned and unaligned sinks such that $\Delta Z_i = Z_{aligned} - Z_{unaligned}$. The sink strengths are a linear function of stress as derived by Savino [25]:

$$Z_{aligned} = z_i \left(1 + \frac{5\sigma(2-\nu)}{2\mu e_o(7-5\nu)} \right), \quad (6)$$

$$Z_{unaligned} = z_i \left(1 - \frac{5\sigma(1+\nu)}{2\mu e_o(7-5\nu)} \right), \quad (7)$$

where μ is the shear modulus, ν is Poisson's ratio, z_i the interstitial bias for the dislocation in a stress-free lattice and e_o is the stress relaxation volume induced by the defect.

Since the defect concentrations are proportional to the dose rate, the dose rate dependence of the creep rate for both SIPA and PAG is also linear. The temperature dependence of the diffusion coefficient D_i cancels the temperature dependence of the reaction rate constants causing both PAG and SIPA to have no temperature dependence. In previous work by Savino [26], [32], the stress induced anisotropic sink strength ΔZ_i was found to be a linear function of stress, making the SIPA mechanism linear in stress dependence and PAG quadratic in stress dependence.

By equating equations (4) and (5), the stress at which the PAG contribution exceeds that of SIPA can be determined:

$$\sigma_{PAG/SIPA} = \frac{Eb}{2} \left(\frac{L}{\pi} \right)^{1/2}, \quad (8)$$

For FM steels, E is around 200GPa, L is typically on the range of $1 \times 10^{10} \text{ cm}^{-2}$ and b around $2 \times 10^{-8} \text{ cm}$ [8]. Then PAG should overtake SIPA at stresses above 112MPa.

However, this calculation assumes the same amount of dislocations participate in both SIPA and PAG. Due to the anisotropic microstructure, more dislocations may prefer to climb or glide depending on the orientation and the stress state of the sample. Therefore, 112MPa is likely not the precise stress upon which PAG overtakes SIPA. However, this calculation does confirm that both mechanisms could be operating under the stress conditions (100-200MPa) of this study.

Both linear and quadratic fits were made to the PLB- and anisotropic loop-adjusted strain rates to determine whether SIPA or PAG was dominating. The fits were forced through zero since irradiation creep goes to zero in the absence of stress. Figure 14 plots the creep strain rates, adjusted for PLB and anisotropic loop strain, as a function of stress. The plot shows that a linear fit goes through the majority of the data well within

the experimental error, suggesting the experimental data can be explained by a linear stress dependence. Similarly, the quadratic fit also falls within the experimental error of the irradiation creep data. The analysis of the stress dependence suggests that both SIPA and PAG are viable irradiation creep mechanisms for FM steels.

The observation of anisotropic dislocation loop distribution would suggest SIPA is indeed operating during irradiation creep but is insufficient to account for majority of the strain rate at high stress. Gelles [33] and Wolfer [34] have both discussed the possibility of dislocation loops growing and interacting with dislocation networks under the influence of stress-induced anisotropic absorption of defects. Woo [5], [6] also expanded on the SIPA mechanism to include the climb of anisotropic dislocation network as a deformation mechanism. Despite the small direct contribution to strain by anisotropic dislocation loops, a combination of the anisotropic microstructure with dislocation climb and glide offer the mechanistic explanation that is most consistent with past observations and with this study. In light of this evidence, a combination of SIPA and PAG is the likely cause of irradiation creep in FM steels.

5. Conclusion

Irradiation creep of alloy T91 exhibited a linear dose rate dependence, power law stress dependence, and negligible temperature dependence. The dose rate dependence and temperature dependence were consistent with neutron irradiation creep under the same conditions. The stress dependence at low stress was also consistent with neutron irradiation creep experiments. The strain rates from proton irradiation creep experiments were found to be $\sim 100\times$ higher than those in reactor experiments due to differences in creep regime, damage morphology, and sample thickness.

The non-zero dose rate dependence and lack of temperature dependence from this experiment suggested that a thermally driven process, such as preferential emission, was not the dominant irradiation creep mechanism. Power law stress dependence provided evidence that power law breakdown dominates creep behavior at high stress.

The anisotropic dislocation loop density observed following proton irradiation creep accounted for only 4%-11% of the total irradiation creep strain. Adjusting the measured creep rates for strain due to the anisotropic loop distribution and power law breakdown showed agreement with the stress dependence predicted by both SIPA and PAG irradiation creep mechanisms. Over the conditions of this study, the likely mechanisms governing irradiation creep in F-M steel is a combination of SIPA and PAG.

Acknowledgements

The authors gratefully acknowledge Ovidiu Toader and Fabian Naab for their assistance in conducting proton irradiations, and Lumin Wang, Kai Sun and Scott Braswell for their assistance in TEM analysis. Micah Hackett for his helpful discussion and insights. The authors also acknowledge the facilities provided by the Michigan Ion Beam Laboratory, the Electron Microbeam Analysis Laboratory at University of Michigan, and Nanotech User Facility at University of Washington. Support for this research was provided by the Department of Energy under award NERI 08-055 DE-FG07-07ID14894.

References

- [1] J. . Matthews and M. . Finnis, “Irradiation Creep Models - An Overview,” *J. Nucl. Mater.*, vol. 159, pp. 257–285, 1988.
- [2] F. A. Garner and D. S. Gelles, “Irradiation Creep Mechanisms: An Experimental Perspective,” *J. Nucl. Mater.*, vol. 159, pp. 286–309, 1988.
- [3] R. . Klueh and D. . Harris, *High Chromium Ferritic Martensitic Materials for Nuclear Applications*. ASTM International, 2001, p. 221.
- [4] C. H. Woo and F. A. Garner, “A SIPA-based theory of irradiation creep in the low swelling rate regime,” *J. Nucl. Mater.*, vol. 194, pp. 1309–1312, 1992.
- [5] C. H. Woo, “Irradiation creep due to climb-induced glide in an anisotropic dislocation structure,” *J. Nucl. Mater.*, vol. 98, pp. 295–305, 1981.
- [6] C. H. Woo, “Effects of an Anisotropic Dislocation Structure on Irradiation due to SIPA,” *J. Nucl. Mater.*, vol. 80, pp. 132–143, 1979.
- [7] L. K. Mansur and T. C. Reiley, “Irradiation creep by dislocation glide enabled by preferred absorption of point defects - theory and experiment,” *J. Nucl. Mater.*, vol. 90, pp. 60–67, 1980.
- [8] L. K. Mansur, “Irradiation creep by climb-enabled glide of dislocations resulting from preferred absorption of point defects,” *Philos. Mag.*, vol. 39, p. 497, 1979.
- [9] C. Xu and G. S. Was, “In situ proton irradiation creep of ferritic-martensitic steel T91,” *J. Nucl. Mater.*, vol. 441, no. 1–3, pp. 681–687, 2013.
- [10] J. F. Ziegler, J. P. Biersack, and M. D. Ziegler, “SRIM 2008.” 2008.
- [11] K. Kroupa, “Dislocation Loops,” in *Theory of Crystal Defects - Proceedings of the Summer School held in Hrazany in September 1964*, 1966, pp. 308–313.
- [12] C. Xu and G. S. Was, “Anisotropic Dislocation Loop Distribution in Alloy T91 during Irradiation Creep,” *J. Nucl. Mater.*, 2014.
- [13] J. Chen, P. Jung, and W. Hoffelner, “Irradiation creep of Candidate Material for Advanced Nuclear Plants,” *J. Nucl. Mater.*, 2013.
- [14] J. Chen, M. a. Pouchon, a. Kimura, P. Jung, and W. Hoffelner, “Irradiation creep and microstructural changes in an advanced ODS ferritic steel during helium implantation under stress,” *J. Nucl. Mater.*, vol. 386–388, pp. 143–146, Apr. 2009.

- [15] B. A. Chin, "Topical Conference on Ferritic Steels for Use in Nuclear Energy Technologies," in *The Metallurgical Society of AIME*, 1984, p. 593.
- [16] M. L. Grossbeck, K. Ehrlich, and C. Wassilew, "An Assessment of tensile, irradiation creep, creep rupture, and fatigue behavior in austenitic stainless steels with emphasis on spectral effects," *J. Nucl. Mater.*, vol. 174, 1990.
- [17] G. S. Was, *Fundamentals of Radiation Materials Science*. Springer, 2007.
- [18] M. B. Toloczko, F. A. Garner, and S. A. Maloy, "In-reactor Creep of Two Heats of T91 (Modified 9Cr-1Mo) from ~ 400 ° C to 600 ° C," in *8th International Workshop on Spallation Materials Technology (IWSMT-8)*, 2006.
- [19] M.B. Toloczko, F.A. Garner, and C.R. Eiholzer, "Irradiation creep and swelling of the US fusion heats of HT9 and 9Cr-1Mo to 208 dpa at ~400°C," *J.Nucl.Mater.*, vol. 212-215, 1994
- [20] R. L. Klueh and a. T. Nelson, "Ferritic/martensitic steels for next-generation reactors," *J. Nucl. Mater.*, vol. 371, no. 1–3, pp. 37–52, Sep. 2007.
- [21] G. Gupta, "Irradiation Creep Behavior of Ferritic-martensitic Alloy T91 by Subgrain Boundary Density Enhancement," University of Michigan, 2007.
- [22] E. M. Haney, F. Dalle, M. Sauzay, L. Vincent, I. Tournié, L. Allais, and B. Fournier, "Macroscopic results of long-term creep on a modified 9Cr–1Mo steel (T91)," *Mater. Sci. Eng. A*, vol. 510–511, pp. 99–103, Jun. 2009.
- [23] M. Kassner, *Fundamentals of Creep in Metals and Alloys*, 2nd ed. Elsevier, 2009.
- [24] K. Kimura, "Creep Deformation, rupture strength, and rupture ductility of grades T/P92 Steels," *ASME Symp. Elev. Temp. Appl. Mater. Foss. Nucl. Petrochemical Ind.*, 2014.
- [25] S. J. Zinkle, J. P. Robertson, and R. L. Klueh, "Thermophysical and Mechanical Properties for Fe-(8-9)%Cr Reduced Activation Steels," 1998.
- [26] E. J. SAVINO and C. N. TOME, "Irradiation Creep by Stress-induced Preferential Attraction due to Anisotropic Diffusion (SIPA-AD)," *J. Nucl. Mater.*, vol. 109, pp. 405–416, 1982.
- [27] G. Gupta and G. S. Was, "Improved Creep Behavior of Ferritic-Martensitic Alloy T91 by Subgrain Boundary Density Enhancement," *Metall. Mater. Trans. A*, vol. 39, no. 1, pp. 150–164, Dec. 2007.
- [28] J. J. Peniston, "The Mechanism of Radiation-induced Segregation in Ferritic-Martensitic Steels," University of Michigan, 2012.

- [29] A. D. Brailsford and L. K. Mansur, "The Effect of Precipitate-Matrix Interface Sinks on the Growth of Voids in the Matrix," *J. Nucl. Mater.*, vol. 104, pp. 1403–1408, 1981.
- [30] B. A. Chin, "An Analysis of the Creep Properties of a 12Cr-1Mo-W-V Steel," in *Topical Conference on Ferritic Alloys for Use in Nuclear Energies Technologies*, 1984.
- [31] A. Kohyama, "Irradiation Creep of Low-Activation Ferritic Steels in FFTF/MOTA," *J. Nucl. Mater.*, vol. 212, pp. 751–754, 1994.
- [32] E. J. Savino, "Stress-induced Preferred Absorption Due to Saddle-point," *J. Nucl. Mater.*, vol. 116, pp. 17–28, 1983.
- [33] D. S. Gelles, "Effects of stress on microstructural evolution during irradiation.," *J. Nucl. Mater.*, vol. 5, pp. 146–161, 1993.
- [34] W. G. Wolfer, "Correlation of radiation creep theory with experimental evidence," *J. Nucl. Mater.*, vol. 90, pp. 175–192, 1980.

TABLES

Table 1. Percent weight composition of T91

	Cr	Mo	Mn	Si	V	Ni	Cu
Wt%	8.37	0.9	0.45	0.28	0.22	0.21	0.17
At%	8.89	0.52	0.45	0.55	0.23	0.20	0.15
	C	Nb	Al	N	P	S	Fe
Wt%	0.1	0.076	0.022	0.048	0.009	0.003	Bal
At%	0.05	0.045	0.045	0.189	0.016	0.005	Bal

Table 2. Strain rate results of irradiation creep experiments.

Sample Name	Temperature (°C)	Stress (MPa)	Dose Rate (dpa/s)	Dose (dpa)	Strain Rate (10^{-9}s^{-1})	Error (10^{-9}s^{-1})
IT450200-A	450	200	1.7×10^{-6}	0.5	12.5	4.46
IT450200-B	450	200	1.7×10^{-6}	0.5	11.5	4.46
IT450180-A	450	180	1.7×10^{-6}	0.5	4.67	1.47
IT450180-B	450	180	1.7×10^{-6}	0.5	5.00	1.47
IT450160-A	450	160	1.7×10^{-6}	0.5	2.7	1.35
IT450140-A	450	140	1.7×10^{-6}	0.5	2.05	1.02
IT450120-A	450	120	1.7×10^{-6}	0.5	1.9	0.95
IT450100-A	450	100	1.7×10^{-6}	1	1.67	0.83
IT450000-A	450	0	1.7×10^{-6}	0.5	N/A	N/A
IT400160-A	400	160	1.4×10^{-6}	0.5	~2.9	1.45
IT500180-A	500	180	5×10^{-6}	0.5	5.78	2.89
IT500160-A	500	160	* 1.5×10^{-6}	0.8	1.78	0.89
IT500160-A	500	160	* 1.7×10^{-6}	0.4	2.33	1.16
IT500160-A	500	160	* 2.4×10^{-6}	0.6	5.08	2.54
TT450200-A	450	200	0	0	1.38	0.69
TT500200-A	500	200	0	0	12.8	5.80
TT500200-B	500	200	0	0	11.5	5.80
IH450160-A	450	160	1.7×10^{-6}	0.5	3.45	1.72
IA450160-A	450	160	1.7×10^{-6}	0.5	5.85	2.92

* Experiment conducted on the same sample at different conditions.

~ Experimental condition difficult to control due to low temperature.

Table 3. Strain due to anisotropic loop density and measured total strain

Sample Name	Loop Anisotropy Strain (%)	Average Loop Size (nm)	Dislocation Loop Density (m^{-3})	Measured Total Strain (%)	Percent of Strain due to Anisotropy (%)
IT450100	0.006	24.3	1.6×10^{21}	0.10	6
IT450180	0.0056	24.9	1.3×10^{21}	0.13	4.3
IT450200	0.0158	42.3	1.0×10^{21}	0.36	4.4
IT500180	0.0067	23.3	1.5×10^{21}	0.06	11.2

Table 4. Input parameters for calculation of point defect concentrations.

Constant Inputs			
Parameter	Value	Units	Notes
T	400-500	°C	Irradiation temperature
σ	100-200	MPa	Applied stress
K_o	$0 - 5 \times 10^{-6}$	dpa/s	Irradiation dose rate
Ω	1.23×10^{-23}	cm ³ /atom	Atomic volume
k	1.38×10^{-23}	Pa/K	Boltzmann constant
e_o	0.8	N/A	Interstitial relaxation volume [31]
ν	0.33	N/A	Poisson's Ratio
μ	75×10^3	MPa	Shear modulus
E	200×10^3	MPa	Elastic modulus
z_i	1.02	N/A	Dislocation bias for interstitials [31]
z_v	1	N/A	Dislocation bias for vacancies [31]
E_{if}	4.6	eV	Interstitial formation energy [27]
E_{vf}	1.7	eV	Vacancy formation energy [27]
E_{im}	0.2	eV	Interstitial migration energy [27]
E_{vm}	0.67	eV	Vacancy migration energy [27]
Measured Inputs: IT 450200			
d	0.45	μm	Sub-grain size
R_{ppt}	0.16	μm	Average precipitate radius [27]
C_{gb}	2.1×10^{13}	cm ⁻³	Sub-grain density
C_{loop}	1.35×10^{10}	cm ⁻²	Dislocation loop line density
$C_{network}$	5.7×10^{10}	cm ⁻²	Dislocation network line density
C_{ppt}	1.16×10^{14}	cm ⁻³	Precipitate density [27]
k_{ppt}	1.4×10^{11}	cm ⁻²	Precipitate sink strength
k_{gb}	7.4×10^{10}	cm ⁻²	Sub-grain sink strength
k_{loop}	6.9×10^{10}	cm ⁻²	Dislocation loop sink strength

k_{network}	4.4×10^{11}	cm^{-2}	Dislocation network sink strength
k_{total}	7.3×10^{11}	cm^{-2}	Total sink strength
$K_{\text{is}} C_{\text{s}}$	2.4×10^6	s^{-1}	Total reaction rate of interstitials to sinks
$K_{\text{vs}} C_{\text{s}}$	6.7×10^5	s^{-1}	Total reaction rate of vacancies to sinks

Figures:

Figure 1. Schematic of irradiation creep sample and samples used for loop orientation analysis.

Figure 2 SRIM result of 3MeV proton beam incident on T91 target. Damage peak occurs around 37 μ m into the sample. Nominal sample thickness of 35 μ m was used to avoid the damage peak and any proton implantation.

Figure 3. Schematic of irradiation creep experimental facility.

Figure 4. Schematic of dislocation loop image method and geometry. The tensile direction is at an arbitrary angle to the <001> image plane, and the angle between the loop normal vector to the tensile axis is defined as θ .

Figure 5. Irradiation creep curve and creep experiment parameters for sample IT450180B. The experiment was conducted at 450°C with an applied stress of 180MPa, and a dose rate of 1.7×10^{-6} dpa/s.

Figure 6. Dose rate dependence of irradiation creep strain rate for IT500160 and linear fit to the data. Sample IT500160 was irradiated at a temperature of 500°C, and a stress of 160MPa.

Figure 7. Temperature dependence of irradiation creep experiments done at 400°C, 450°C, and 500°C at applied stress of 160MPa and dose rate of 1.7×10^{-6} dpa/s.

Figure 8. Stress dependence of irradiation creep rates of T91 at 450°C, 1.7×10^{-6} dpa/s and the best line fits to the data.

Figure 9. TEM image of a_0 <100> edge-on loops with g =<110> on the <100> zone axis. a) IT450100, b) IT450180, c) IT500180, d) IT450200.

Figure 10. Loop anisotropy plot of the irradiation creep experiments for a) IT450100, b) IT450180, c) IT500180, d) IT450200 (taken from [12]).

Figure 11. Neutron irradiation creep data of T91 by Tolockzo [18] plotted as a function of irradiation temperature.

Figure 12. Comparison of proton irradiation creep and neutron irradiation creep rates [18] adjusted for dose rate.

Figure 13. Calculation of point defect concentration as a function of temperature with measured sink densities of IT450200 shown in Table 4.

Figure 14. Stress dependence of proton irradiation creep rates without contribution from anisotropic dislocation loops and power law breakdown with linear and quadratic fits.

Fig 1A

Fig 2

Fig 3

Fig 4

Fig 5

Fig 6

Fig 7

Fig 8

Fig9

Fig 10

Fig 11

Fig 12

Fig 13

Fig 14