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Irradiation creep of oxide dispersion strengthened (ODS) steels for advanced nuclear applications

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A B S T R A C T

Ferritic oxide dispersion strengthened steels with different microstructure were in-beam creep tested in a temperature range from 300 °C to 500 °C. Irradiation was by He-ions. Elongation was determined as a function of stress and irradiation damage rate. Damage was investigated by transmission electron microscopy. A thorough analysis of the loops developing during irradiation creep did not show any dependence of orientation or size on the direction of the applied stress. At 400 °C radiation induced segregation was found (most probably an iron aluminide) which had no effect on irradiation creep. No pronounced influence of microstructure or dispersoid size on the irradiation creep behavior was detected. Irradiation creep compliance of PM2000 with dispersoids of about 30 nm diameter were found to differ little from material with dispersoids of only 2–3 nm diameter. This is in contrast to thermal creep where dislocation–obstacle interactions are extremely important. An assessment of the technical relevance of irradiation creep in advanced nuclear systems is presented.

1. Introduction

Oxide dispersion strengthened (ODS) steels are candidate materials for a variety of nuclear applications including advanced cladding or structural parts. The main interest in this class of materials comes from its very good thermal creep properties, making them suitable for high temperature applications. ODS materials are available with austenitic (primarily nickel-based), ferritic, and ferritic–martensitic matrices. Ferritic–martensitic steels with 9–12% Cr are considered for elevated temperature applications, depending on the stability of martensite. The ferritic grades (also for high temperature applications) usually have Cr-contents of up 20% and sometimes they contain aluminum, providing good oxidation resistance. In our investigations we studied ferritic ODS steels containing about 20% chromium. Main work was done with the then commercially available PM2000 [1] (Plansee/Austria) in the annealed condition. A fine grained Japanese ferritic ODS steel (19 Cr ODS [2]) with nano-sized dispersoids was also investigated. Thermal creep in metals occurs at temperatures above about 0.35 T_m (where T_m is the melting point of the alloy). Deformation under static load occurs at much lower temperatures when simultaneous irradiation occurs, and is known as irradiation creep. Technically, this creep is important only at temperatures below 550 °C. Irradiation creep could become a design issue for components being exposed during operation only to moderate temperature or during transient conditions like start-up for high temperature plants. Irradiation creep has been studied in the past under helium implantation and neutron irradiation mainly with respect to fusion and fast fission reactors for austenitic, ferritic, and ferritic–martensitic steels [3–5]. Differences in irradiation creep behavior between neutron irradiation and helium implantation cannot be excluded. For relative comparison between different microstructures of the same type, these effects are not expected to play a significant role. The paper summarizes irradiation creep data and related microstructural findings. The relevance of these findings for design of advanced nuclear plants like gas cooled reactors or advanced fast spectrum reactor concepts is discussed.

2. Materials and testing conditions

The investigations were performed with two ferritic ODS steels. Typical dispersoid distributions and microstructural parameters are shown in Fig. 1 and Table 1. The number densities of dispersoids were $5.1 \times 10^{20}/m^3$ in PM2000 and $1.2 \times 10^{24}/m^3$ in 19 Cr ODS. The micrographs show size and distribution of the dispersoids. The grain sizes vary from very large crystals in one direction to sizes below 500 nm. The annealed material was supplied by Plansee. It should be mentioned that this material is no longer produced by this company. The 19 Cr ODS was supplied by A. Kimura (Kyoto University) [2].

In situ irradiation creep under He-implantation was performed at the compact cyclotron of Forschungszentrum Juelich. Dog-bone samples with a gage length of 10 mm, a width of 2 mm and a thickness of 0.1 mm were used. Creep strains were determined as average of two linear variable differential transformer (LVDT) signals of

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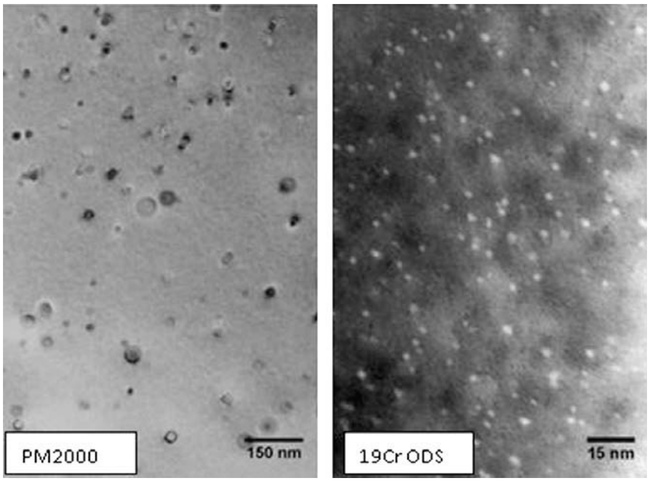

Fig. 1. Dispersoid sizes, distributions and microstructural features of the materials investigated. Note that the two micrographs are at different magnifications.

two ‘rod-pipe’-type extensometers. The resolution of creep strain measurements was 3×10^{-6}. More details of the experimental set up are described in [6]. With 24 MeV $^4He^{++}$ ions passing through a magnet scanning system and a degrader wheel with 24 Al-foils of variable thicknesses, 0.1 mm thick samples were 3D-homogeneously irradiated under constant uniaxial stress. The dose levels reached per creep test at each temperature were about 0.2 dpa. The damage distribution with depth can be seen from the results of TRIM calculations shown in Fig. 2.

More details about the experimental conditions can be found in literature [7]. Thermal creep tests were performed outside of the irradiation facility for comparison, with similar equipment using the same sample geometry.

3. Results

Microstructural changes as a result of irradiation creep were analyzed for PM2000 by transmission electron microscopy. Loops of types ½ <1 1 1> (1 1 1) and <1 0 0> (1 0 0) were detected at 300 and 400 °C. At 500 °C a dislocation network was observed in addition to the loops. Average loop diameters and loop densities are shown in Table 2.

Loop diameters differed with temperature by one order of magnitude and loop densities by three orders of magnitude. The most important result was, however, that the loops were circular and no correlation between loop orientation or geometry and direction of the applied stress was found. This is in contrast to the current understanding of irradiation creep as described in more detail in [8]. Another interesting finding was the development of a radiation induced ordered phase (most probably Fe_3Al) at 400 °C in PM2000. No such phase is expected for 19 Cr ODS because it does not contain aluminum (see Table 1).

The average particle size in PM2000 was determined to be 3.6 nm; the average density of the particles was $3 \times 10^{-23}\ m^{-3}$. Two explanations are possible for this observation [5]. One explanation could be that irradiation enhanced diffusion promotes phase equilibria at low temperatures where thermal diffusion becomes exceedingly low. The other explanation would be that the presence of non-equilibrium point defects during irradiation affects the free energies of phases thereby shifting the solubility.

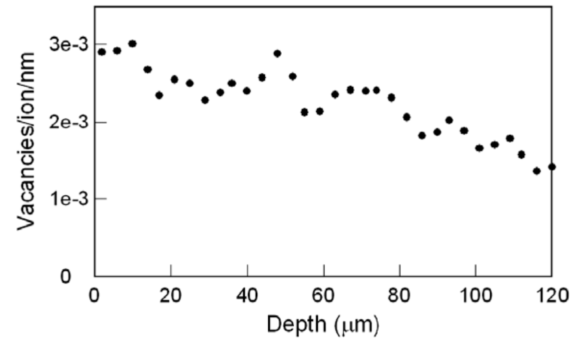

Fig. 2. The dependence of displacement damage from penetration depth in the irradiation creep samples (TRIM calculations).

Table 2
Average loop diameters and loop densities determined in irradiation creep samples for PM2000. At 500 °C a dislocation network was also observed.

Temperature (°C)	Average loop diameter (nm)	Average loop density (/m^3)
300	4.6	4.80E + 23
400	62.3	4.60E + 21
500	31	3.00E + 20

Strain rates were determined from irradiation creep curves which showed a steady state creep behavior similar to thermal creep. In Fig. 3 the ratio between strain rates and dose rates is plotted as a function of the applied stress. Some scatter of the data was observed which is usual for mechanical properties of small samples. However, no significant differences between the two materials PM2000 and 19 Cr ODS were found. The slopes of the creep curves (called irradiation creep compliance, C) are listed in Table 3 (see also [9]). A plot of the compliances as a function of $1/T$ is shown in Fig. 4.

In the temperature range investigated only a weak temperature dependence of the irradiation creep compliance, C, was found and was determined to be:

$$C(T) = 0.0003 \cdot e^{-2335/T}. \quad (1)$$

Determination of compliances from published results on neutron irradiated pressurized tubes of different ferritic and ferritic–martensitic alloys [5] revealed the same temperature dependence (slope of curve) at temperatures of 500 °C or lower. The compliances measured in the present work were about a factor of ten higher than expected for ferritic materials based on results of creep experiments under neutron irradiation with total doses higher than 10 dpa [3,5]. This can be explained as follows. Evaluation of irradiation creep rates for austenitic stainless steels [10] indicate a sharp drop of irradiation creep compliances at total doses between 0 and 2 dpa. For higher doses the compliances are constant. For a total dose of 0.2 dpa (our experiments) about one order of magnitude

Table 1
Chemical composition and microstructure of the ODS steels investigated.

Material	Dispersoid diameter (nm)	Grain size (μm)^3	Cr (wt%)	Al (wt%)	Ti (wt%)	W (wt%)	Y_2O_3 (wt%)	Fe
PM2000 annealed	28.0	1000 × 1000 × (>10,000)	19.0	5.5	0.5	–	0.5	Bal.
19 Cr ODS	2.1	0.2 × 0.2 × 0.7	18.4	–	0.3	0.3	0.4	Bal.

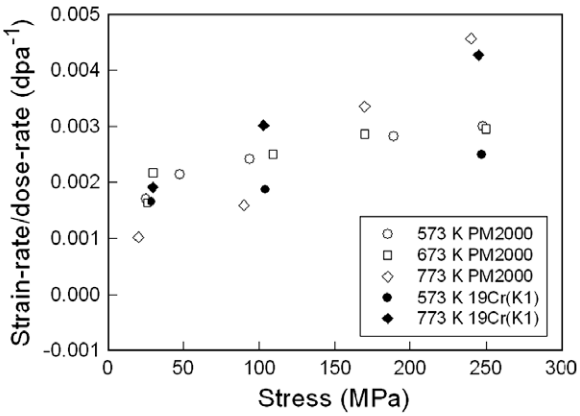

Fig. 3. Ratio of strain rate and dose rate as a function of applied stress for two ferritic ODS steels measured at 300–500 °C.

Table 3
Irradiation creep compliances in $\frac{\varepsilon}{dpa}$ MPa at different temperatures for the two steels

Temperature (°C)	PM2000 annealed ($\times 10^{-6}$)	ODS 19 Cr ($\times 10^{-6}$)
300	5.7	4.0
400	5.7	–
500	18	11

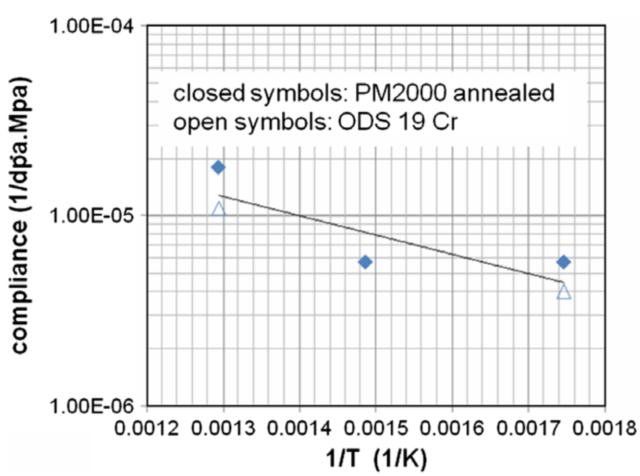

Fig. 4. Irradiation creep compliance $\frac{\varepsilon}{dpa}$ MPa as a function of the inverse temperature (in 1/K).

higher compliance is expected than for total doses above 2 dpa. This is fully consistent with our findings. With respect to irradiation creep there is no difference between light ions and neutrons reported in [10].

4. Discussion

The investigated materials belong to the class of ferritic ODS steels containing about 20% Cr. However, microstructures were considerably different. The grain sizes varied from 200 nm up to almost single crystalline. Only PM2000 contained aluminum. Dispersoid diameters differed by more than an order of magnitude. Size and density of loops were determined for irradiated PM2000. They varied by orders of magnitudes in a temperature range from 300 to 500 °C at a dose of 0.7 dpa . A radiation induced phase was formed in PM2000 at 400 °C (but not at the other temperatures). Although each of these features is expected to have an influence on strength and thermal creep properties, the surprising finding was that no significant effect on irradiation creep compliance could be found in the temperature range investigated. In particular, the fact that one order of magnitude difference in dispersoid size has no significant influence is in contrast to thermal creep results [11]. These findings indicate that irradiation creep seems to be decoupled from dislocation–obstacle interactions. It might be speculated that irradiation creep is pure matrix behavior. Irradiation creep clearly has an effect on the accumulation of strain or on the relaxation of stress in components operating under such conditions. As a result of the detected independence of irradiation creep on microstructure we will confine our considerations to annealed PM2000. Both thermal creep and irradiation creep will be considered. Due to lacking experimental data only a preliminary assessment can be developed, describing only the most important consequences. For the determination of thermal creep we shall base our considerations on stress rupture curves and the Monkman–Grant relationship between secondary creep rate and stress rupture life, which is a well accepted technical procedure for such assessments. A parametrization of stress rupture curves based on existing literature data for PM2000 was given by the authors at the HTR2006 conference [12] using the relationship in Eq. (2)

$$\log 10(t_R) = T \cdot (A \cdot \log 10(\sigma) + B\sigma + C) + D, \quad (2)$$

where t_R = time to rupture, σ = stress, T = absolute temperature, and A,B,C,D are fitting parameters.

The dependence of strain rate on creep rupture times was assessed in terms of the Monkman–Grant relationship established with limited creep data on PM2000 annealed. The results are shown compared to literature data for the ferritic martensitic mod 9 Cr steel (grade 91) [13] in Fig. 5. The slight deviation of grade 91 from the Monkman–Grant relationship at long rupture lives is neglected for our considerations here.

Although there is not enough data available for a thorough evaluation of PM2000 the approach looks plausible and applicable for a first assessment. Slopes of the T91 line and the PM2000 line are comparable and a clear influence of the dispersoids can be seen in the significantly lower strain rates at given stress rupture lives.

Using the stress rupture parametrizations (Eq. (2)), the Monkman–Grant relationships and an average irradiation creep compliance determined according to (Eq. (1)) relaxation curves for thermal and irradiation creep were calculated for a temperature of 500 °C at a stress level of 150 MPa, shown in Fig. 6. A commercially available program (STELLA 9.1) [14] was used for this numerical integration. In agreement with GEN IV specifications a relevant time period of 60 years was chosen. The dpa-levels accumulated over the whole period cover VHTR structures (0.1–10 dpa). Fast reactors and fusion would be much higher (60–200 dpa). It can

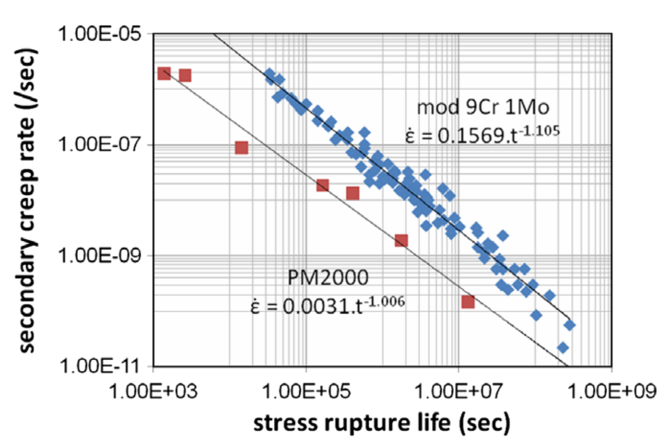

Fig. 5. Monkman–Grant plots of mod 9 Cr (after [11]) and PM2000 annealed.

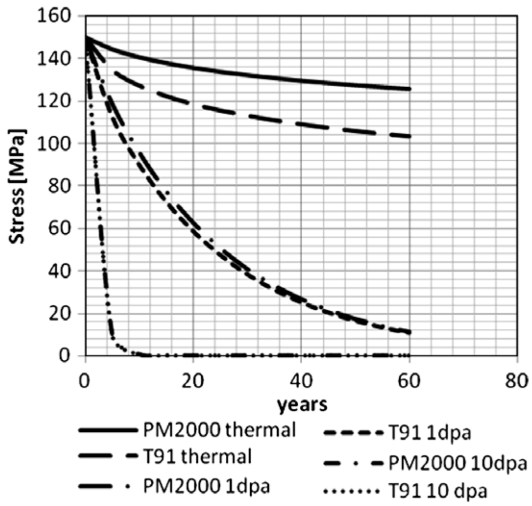

Fig. 6. Stress relaxation of 150 MPa over 60 years at 500 °C calculated with the irradiaton creep compliance determined under He-implantation. For 10 dpa no difference between PM2000 and T91 exists.

be seen that even for 1 dpa, irradiation creep becomes the only important mechanism under the present irradiation conditions. Assuming that neutron irradiation to higher doses would reduce the effect by about one order of magnitude (as discussed above) the influence of irradiation creep would be shifted accordingly. Concerning advanced nuclear application the implications would be minor because for all doses $\geqslant$10 dpa in 60 years irradiation creep would remain the dominant mechanism. As no significant influence of microstructure on irradiation creep was found these curves are expected to be valid for all ferritic and ferritic martensitic ODS steels. Although ODS materials are also expected to operate at higher temperatures (where irradiation creep is of little or no importance) significant irradiation creep can be expected during transient conditions like start-up. In such a situation most probably the higher compliances become important because of only small total dose per transient. It should also be taken into consideration that irradiation creep shows a rather weak temperature dependence which means that even below 500 °C significant irradiation creep is expected. Taking the scatter of the data and the limited amount of available data into consideration the relaxation curves can act as guidelines but they can currently not be considered as design curves for irradiation creep. For that purpose not only more material data but also clear design needs (stresses, temperatures, doses, etc.,) would be required.

5. Conclusions

Irradiation creep of ferritic 20% Cr ODS alloys was investigated in the temperature range 300–500 °C. Microstructural investigations revealed no relationship between loop size or loop shape and the applied stress. This behavior, which is not predicted by irradiation creep models for bcc matrix alloys, needs further investigation. The formation of a radiation induced FeAl intermetallic phase in PM2000 at 400 °C had no effect on the irradiation creep behavior. The most interesting finding was that size and distribution of dispersoids did not show a significant influence on the irradiation creep behavior. It might be inferred that irradiation creep is a pure matrix phenomenon. Irradiation creep properties were therefore discussed using PM2000 data only. To assess the importance of thermal creep relative to irradiation creep, the thermal creep properties of PM2000 were estimated based on literature results and our own data. Irradiation creep does not show strong temperature dependence. For temperatures up to 500 °C creep and relaxation behavior of components are dominated by irradiation creep. Even under the assumption that (based on literature) the expected irradiation creep compliances under neutron irradiation to higher doses would be lower than under the irradiation conditions of our investigation, irradiation creep can be considered of technical relevance in advanced fast fission reactors and fusion plants.

Acknowledgements

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