Low-Stress Creep Deformation in Long-Term Aged Ferritic Heat-Resistant Steel

Shigeto Yamasaki1,*, Masatoshi Mitsuhara2, Ken-ichi Ikeda2, Satoshi Hata2 and Hideharu Nakashima2

1Interdisciplinary Graduate School of Engineering Sciences, Kyushu University, Kasuga 816-8580, Japan
2Faculty of Engineering Sciences, Kyushu University, Kasuga 816-8580, Japan

The transition of the creep deformation mechanism in the low-stress region of Grade P92 high Cr ferritic heat-resistant steel was investigated by a helicoid spring creep test. Specifically, the effect of variation in the microstructure of steel on creep deformation behavior was evaluated by subjecting samples to thermal aging for 1000, 3000, 5000, and 10000 h at 700°C over a wide stress range. In addition, stress exponents were determined from the stress dependence of the minimum strain rate in the creep curves up to 270 ks. The transition of the creep mechanism was indicated when the stress exponent decreased from 4 in the high-stress region to 1 in the low-stress region below 40 MPa. A quantitative evaluation of the microstructure of a tempered martensite sample, including the determination of the amount of dissolved Mo and W, dispersion state of the precipitates, and length of the grain boundaries per unit area, was also carried out. Furthermore, the change in the minimum strain rate was evaluated as a function of the microstructural changes that accompanied thermal aging. It was found that the change in the strain rate was the most affected by the fineness of the martensitic lath structure in the high-stress region and by the dispersion density of M23C6 precipitates in the low-stress region. Based on these results, it was concluded that the microstructural parameter that most affects creep deformation behavior differs depending on the stress region due to the difference in the creep mechanism. [doi:10.2320/matertrans.M2013427]

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1. Introduction

High Cr ferritic heat-resistant steel is used for the material of components of thermal power plants, which are exposed to elevated temperatures for extensive periods. Under these conditions, microstructural changes due to creep deformation occur in the material. Several types of microstructural changes are caused by creep deformation, such as the growth of precipitates at elevated temperatures, variation in the dislocation density due to the introduction of strain into the material, and coarsening of the martensitic lath structure caused by both the introduction of strain and extended exposure to elevated temperatures. In high Cr ferritic heat-resistant steel, it is known that predictions based on the results of creep tests under high-stress conditions using the time-temperature parameter method overestimate creep strength under low-stress conditions.1-4) This phenomenon is believed to be due to a decrease in the number of obstacles that prevent dislocation movement by the annihilation of subgrain boundaries. This reduction occurs as a result of the coarsening of the precipitates in steel, which is caused by creep deformation due to exposure at elevated temperatures for long durations.5,6) Therefore, studies of the creep deformation of high Cr ferritic heat-resistant steels tend to be focused on revealing the microstructural changes that occur during creep tests or during actual operation over an extended time period. Several researchers have recently reported, however, that according to the results of long-term creep rupture tests,7-9) the stress dependence of the minimum strain rate of ferritic heat-resistant steel changes in the low-stress region. Because the dependence of the stress on the minimum strain rate corresponds to the creep mechanism, the change in stress dependence indicates that the degradation of creep rupture strength in the low-stress region is caused by the transition of the creep mechanism. That is, the degradation of creep strength in the low-stress region may be caused by one or two completely different physical phenomena of the transition of the creep deformation mechanism or a change in the microstructure. However, because the minimum strain rate, which is evaluated via long-term creep tests, includes the influence of the microstructural changes during the creep test, it is difficult to separately determine the extent of the contribution of the microstructural changes and the transition of the creep mechanism to the creep deformation behavior. Among the microstructural changes that occur during creep deformation, those caused by long-term exposure at elevated temperatures can be reproduced in long-term thermal aging tests.

Herein, we report the results of our evaluation of the influence of the microstructural changes resulting from long-term aging on the creep behavior in pre-aged ferritic heat-resistant steels. We also describe the results of our investigation of the transition of the creep mechanism of these steels during short-duration, low-stress creep tests.

2. Experimental

2.1 Material and heat treatment

Grade P92 ferritic heat-resistant steel was used in the experiments, and its chemical composition is shown in Table 1. The steel was first normalized at 1080°C for 1 h and tempered at 760°C for 1 h (unaged) and then thermally aged at 700°C for 1000, 3000, 5000, or 10000 h.

2.2 Microstructural observation

The Microstructure in the prepared samples were observed using a scanning electron microscope (SEM), and back-scattered electron (BSE) imaging was employed to observe the precipitates in the steel samples. Energy dispersive X-ray spectroscopy (EDS) and electron backscattered diffraction
(EBSD) analysis were also carried out to evaluate the microstructural changes due to aging. For SEM observation, 5 mm square samples were cut from the bulk material and ground using emery paper. The ground samples were then polished with diamond powder, etched using Vilella’s reagent, and subsequently polished using a colloidal suspension of silica with a diameter of 50 nm. The quantitative evaluation of the precipitate particles was performed by measuring 150–700 particles in four fields of view for a single martensitic packet grain. The types of precipitates were identified on the basis of the EDS analyses results and their brightness in SEM-BSE images. A single EBSD measurement was performed for a 200 μm × 200 μm area at 500 nm intervals, and crystallographic orientation distribution maps were obtained for four fields of view. The data were analyzed using TSL OIM Analysis Ver. 6 software, and crystallographic orientation distribution maps were described with a hexagonal lattice pattern.

2.3 Creep test
The helicoid spring creep test method was used for the evaluation of low-stress creep behavior. Helicoid spring creep specimens were fabricated from a cylindrical ingot of the unaged and the aged samples by machining. Outer diameter of the specimen is 8.5 mm, and its inner diameter is 6.8 mm. A wire part of the helicoid specimen has a rectangular cross section. A long side of the rectangular cross section is parallel to a loading direction and its length is 1.70 mm. A short side of the rectangular cross section is vertical to a loading direction and its length is 0.85 mm. The displacement of coil pitch distance was measured using an optical micrometer with a displacement resolution of 150 nm. And, the displacement was converted to shear strain by assuming torsion of the wire part. The creep test temperature was 650°C, and shear stresses of 5.8, 17, 29, 40, 58, and 75 MPa were applied. These applied shear stress values were calculated as the maximum shear stress generated at the midpoint of the long side in the rectangular cross section by loading. Every creep test was interrupted at 270 ks in a transient creep region to minimize the microstructural evolution during the test. One helicoid spring specimen for each aging condition was firstly crept under the shear stress of 5.8 MPa. After interrupting the test, the same specimen was crept once again under the shear stress of 17 MPa. In this way, six times of creep tests were carried out using one spring specimen in order of stress level from lowest to highest. Note that the creep deformation behavior such as a minimum strain rate in a creep curve up to 270 ks obtained from these multiple creep tests is hardly discriminated from that obtained with a single stress condition.

3. Results and Discussion
3.1 Microstructural changes due to thermal aging
Low-magnification SEM-BSE images of the unaged and aged steels are shown in Figs. 1(a)–1(e). The unaged steel
showed a typical tempered martensitic lath structure including packet grains, block grains, and martensite laths or subgrains in prior austenite grains. A higher-magnification image of the sample aged for 1000 h is shown in Fig. 1(f). In this image, at least three types of precipitates of different brightness were observed. EDS analysis revealed that the brightest precipitates were rich in Mo and W, the intermediate brightness precipitates were rich in Cr, and the darkest brightest precipitates were rich in V. According to the literature, the precipitates can be assigned as Laves phase, M23C6, and MX precipitates, respectively. Many M23C6 particles can be mainly observed on the grain boundaries, while the fine MX particles exist both in martensitic lath grains and on lath boundaries.

After aging for 1000 h at 700°C, coarse Laves phases appeared on the prior austenite boundaries, and these phases coarsened with increasing aging time, while their number density decreased. Because the Laves phases contain large amounts of Mo and W, which act as solute strengthening elements in ferritic heat-resistant steel, the increase in the amount of precipitated Laves phase particles led to a decrease in the amount of dissolved Mo and W in the matrix. The variation in the amount of dissolved Mo and W with aging time is shown in Fig. 2. According to Fig. 2, the amount of dissolved Mo decreased slowly up to 3000 h and then remained nearly constant beyond 3000 h. The amount of dissolved W, on the other hand, decreased relatively faster than Mo and reached a constant value at 1000 h. This decrease in the amount of dissolved Mo and W corresponds well to the increase in number of Laves phases observed in the SEM-BSE images (Fig. 1(a)–1(e)). Because the amount of dissolved Mo and W in the matrix became nearly constant after 3000 h, it can be concluded that precipitation of the Laves phases was complete by 3000 h, and only the coarsening of the Laves phase particles occurred beyond this point.

The SEM-BSE images of the precipitates in a single martensitic packet grain are shown in Fig. 3; both medium- bright M23C6 particles and darker MX particles were observed in the field of view. The area fraction of each precipitate (a), the average diameter of the precipitated particles (b), and the average surface spacing of the M23C6 and MX particles (c) are plotted against the aging time in Figs. 4(a)–4(c), respectively. The average surface spacing of the particles $\lambda$ was determined using the following equation:

$$\lambda = 1.25N^{-\frac{1}{2}} - \tilde{d},$$

where $N$ is the number of each particle in the unit area, and $\tilde{d}$ is the average diameter of each particle. The area fraction of MX particles remained nearly unchanged, while the average diameter of these particles slightly coarsened. In contrast, the area fraction of M23C6 particles increased at 1000 h and then decreased after 1000 h, while the average diameter of these particles reached a minimum value at 1000 h and then coarsened beyond 1000 h. Therefore, it can be concluded that the precipitation of MX particles was complete at after tempering and the particles were coarsened monotonically with increasing aging time. The precipitation of M23C6 particles, on the other hand, occurred during aging up to 1000 h and then the particles were coarsened beyond 1000 h.

The crystallographic orientation distribution maps of the unaged and aged steels obtained by EBSD analysis are shown in Figs. 5(a)–5(e). From the figures, coarsening of the martensitic packet grains and block grains occurred during aging at 700°C. The variation in high-angle boundary length and lath boundary length per unit area obtained from the crystallographic orientation distribution maps with increasing aging time is shown in Fig. 6. High-angle boundaries were defined as boundaries with misorientations greater than 15°, and lath boundaries were defined as boundaries with misorientations of 1 to 5°, given that the misorientation of lath boundaries has been reported to range from 0.5 to 5°.

![Fig. 2 Amount of dissolved Mo and W in the ferrite matrix in the unaged and aged steels.](image)

![Fig. 3 SEM-BSE images of the M23C6 and MX particles in a single martensitic packet grain in the (a) unaged steel and in steels aged at 700°C for (b) 1000 h, (c) 5000 h, and (d) 10000 h.](image)
Fig. 4 Change in the (a) area fraction, (b) average diameter, and (c) average surface spacing of the M$_2$C$_6$ and MX particles in the unaged and aged steels.

Fig. 5 Crystal orientation distribution maps for the (a) unaged steel and steels aged at 700°C for (b) 1000 h, (c) 3000 h, (d) 5000 h, and (e) 10000 h.
EBSD is approximately 0.4°. In Fig. 8, a larger value for the boundary length per unit area indicates a finer grain size or martensitic lath structure. Both the length of the high-angle boundary and the lath boundary per unit area decreased as the aging time increased up to 5000 h. This decreasing of the boundary length means the coarsening of the packet, block, and lath grains. In addition, because the boundary length per unit area did not change from 5000 to 10000 h, it was concluded that the coarsening of the grains was saturated at 5000 h.

3.2 Creep deformation behavior

The creep curves are shown in Fig. 7. The elastic strain calculated as 60.3 GPa of the shear modulus of the unaged steel at 650°C is also shown in the figure. According to Fig. 1, in all specimen and all test conditions, shear strain gradually increases as time under a constant stress. Then, the minimum strain rate \( \gamma_m \) in the obtained creep curves was determined by fitting the following equation\textsuperscript{15)} for each creep curve:

\[
\gamma = \gamma_i + \gamma_a[1 - \exp(-\beta_a t)] + \gamma_p[1 - \exp(-\beta_p t)] + \gamma_{at} t,
\]

where \( \gamma \) is the shear strain, \( \gamma_i \) is the instantaneous strain, \( \gamma_a \) and \( \gamma_p \) are the saturation values of shear strain of anelastic deformation and transient creep, respectively, \( \beta_a \) and \( \beta_p \) are the rate constants for anelastic deformation and transient creep, respectively, and \( t \) is the creep time. Because the creep tests in present study were interrupted in short duration, it was expected that a contribution of anelastic deformation in the obtained creep curves would be relatively large. Based on the assumption, we used the above eq. (2) for fitting the experimental creep curves with taking the anelastic deformation into account. Furthermore, for the same reason as it mentioned before, the minimum strain rate in its rigorous meaning is not obtained by the present creep test method, and an evaluated \( \gamma_m \) almost coincided with the strain rate at just before the end of a creep test. Therefore, we should discuss the \( \gamma_m \) as a strain rate at the same interruption time in a transient creep stage. However, the term, \( \gamma_m \), in the present study is referred as “the minimum strain rate” for convenience. Each constant was determined such that the correlation coefficient between the creep curve and the fitting curve was 0.99 or more. The dependence of the minimum strain rate on the shear stress for each sample is shown in Fig. 8. A power law relationship between the applied shear stress and the minimum strain rate was observed. This relationship is expressed as \( \gamma_m \propto \tau^n \), and the stress exponent value changes at approximately 40 MPa. In the high-stress region, the value of the stress exponent was approximately 4, while in the low-stress region below 40 MPa, it was about unity. Kimura et al.\textsuperscript{8)} revealed that the stress exponent changes at a boundary stress of approximately 90 MPa in a tensile stress for Grade P92 steel at 650°C. The stress exponents reported by them were approximately 13 and 5 above and below 90 MPa, respectively. A tensile stress of 90 MPa can be converted to a shear stress of 52 MPa using the von Mises criterion. Therefore, the boundary stress for the stress dependence of the minimum strain rate obtained by Kimura et al.\textsuperscript{8)} is similar to that obtained in this study. In contrast, the stress exponent values in each stress region were quite different in both studies. As it was mentioned in the introduction, the value of the stress exponent corresponds to the creep mechanism. However, in the case of the helicoid spring creep test, when a strain rate distribution across the rectangular cross section of the coil wire part is not proportional to the distance from the center of the torsion due to the occurrence of a plastic deformation, stress redistribution occurs in the rectangular cross section. In such a case, the maximum shear stress during creep deformation decreases in comparison to the stress evaluated in an elastic situation, and thus, the apparent stress exponent is underestimated under these conditions.\textsuperscript{16)} Therefore, it should be considered that the stress exponents obtained in this study is underestimated. Moreover, because the creep tests in this study were interrupted at 270 ks, it may lead to the underestimation of the values of the stress exponent. On the other hand, the stress exponents evaluated via long-term creep-rupture tests\textsuperscript{7-9)} do not directly reflect the creep deformation mechanism either, because these stress exponents include the influence of the microstructural changes that occur during the creep tests. Consequently, the creep deformation mechanisms in high and low stress regions should not be identified by the apparent stress exponent values. Note, however, that the values in respective creep tests are varied at the similar stress level. In addition, the influence of microstructural changes on the creep deformation behavior can be negligible in the present study because of the short duration of a creep test, 270 ks. For these reasons, we conclude that the change of the stress exponent occurred by the transition of the creep deformation mechanism. Furthermore, in the higher stress region than boundary stress of 40 MPa, the unaged steel had the smallest shear strain in the creep curves shown in Fig. 7, but in less than 40 MPa, the sample aged for 1000 h had the smallest shear strain. This result indicates that if the creep deformation mechanism is different in the different stress regions, the microstructure which has the strongest effect on the creep deformation behavior is also different. Hence, by considering the relationship between the creep deformation behavior and
3.3 Microstructural parameters affecting creep deformation behavior

The minimum strain rates \( \dot{\gamma}_m \) determined by the helicoid spring creep tests up to 270 ks at each applied shear stress are plotted against the aging time in Fig. 9. Above 58 MPa, the \( \dot{\gamma}_m \) value for the unaged steel was the lowest of all of the sample materials. In addition, the \( \dot{\gamma}_m \) value increased up to 5000 h of aging time and then slightly decreased for the sample aged for 10000 h. In contrast, the \( \dot{\gamma}_m \) value for the unaged steel was the highest below 29 MPa, while the lowest \( \dot{\gamma}_m \) value was observed for the steel sample aged for 1000 h, and this value slightly increased beyond 1000 h. Furthermore, the \( \dot{\gamma}_m \) value at 40 MPa was in between those above 58 MPa and below 29 MPa. These results indicate that the relationship between the \( \dot{\gamma}_m \) value and the microstructure of the sample materials varied depending on the applied shear stress. In the high-stress region above 58 MPa, the \( \dot{\gamma}_m \) value was higher when the high-angle boundary length or the lath boundary length became shorter. Because both the high-angle boundary length and lath boundary length per unit area

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Fig. 7 Creep curves of the unaged and aged steels at 650°C.
decreased with increasing aging time (Fig. 6), it is difficult to separately estimate the influence of the variation of these values on the $\dot{\gamma}_m$ value. However, particularly in the high-stress region, it is known that the fine martensitic lath structure is the microstructure to the highest extent in terms of decreasing the strain rate of creep deformation.\textsuperscript{5,6) Therefore, it is reasonable to conclude that the $\dot{\gamma}_m$ value of a material that has a fine martensitic lath structure will be low in the high-stress region.

On the other hand, there was no correlation between the fineness of the martensitic lath structure and the $\dot{\gamma}_m$ value in the low-stress region below 29 MPa. In the low-stress region, the $\dot{\gamma}_m$ value for the unaged steel was the highest. However, the $\dot{\gamma}_m$ value became the lowest in the steel aged for 1000 h and then increased again in the materials aged for more than 3000 h. These changes in the $\dot{\gamma}_m$ value did correspond well with the changes in the average surface spacing of M\textsubscript{23}C\textsubscript{6} precipitates (Fig. 4(c)). That is, the $\dot{\gamma}_m$ value correlated the best with the martensitic lath structure in the high-stress region, but with the average surface spacing of the M\textsubscript{23}C\textsubscript{6} precipitates in the low-stress region. It should be noted that, because the amount of dissolved Mo and W was nearly constant after 3000 h of aging time, it was concluded that the dissolved amount of these solute strengthening elements did not strongly affect the $\dot{\gamma}_m$ value in either stress region.

The boundary value of the stress at which the change in the microstructure most correlated to the $\dot{\gamma}_m$ value corresponded to the boundary stress of the stress exponent shown in Fig. 8. Consequently, it was considered that, because the creep mechanism changed at the 40 MPa, the microstructure that most correlated with creep deformation behavior also changed. Because there was a strong correlation between the lath boundary length per unit area and the $\dot{\gamma}_m$ value, the martensitic lath structure was concluded to be the main obstacle against dislocation motion in the high-stress region above 40 MPa. Thus, the predominant creep mechanism in the high-stress region was determined to be dislocation creep.\textsuperscript{17) On the other hand, in the low-stress region below 40 MPa, there was a strong correlation between the dispersion state of the M\textsubscript{23}C\textsubscript{6} particles and the $\dot{\gamma}_m$ value, and the correlation between the martensitic lath structure and the $\dot{\gamma}_m$ value disappeared. Kloc \textit{et al.}\textsuperscript{18}) carried out helicoid spring creep tests with respect to a heat-resistant steel similar to the sample material used in this study. Because the stress exponent evaluated below 35 MPa was 1, they concluded that the predominant creep mechanism was viscous creep, such as Nabarro-Herring\textsuperscript{19}) creep or Coble creep.\textsuperscript{20)} While the stress exponent value reported by Kloc \textit{et al.} is the same as that obtained in this study, it is difficult to evaluate the true stress exponent value using the helicoid spring creep test, and thus inferring the creep mechanism based on the results obtained using this test method is not appropriate. In addition, according to the constitutive equations of the viscous creep mechanism, the minimum strain rate should be proportional to the square or the cube of the inverse of the grain size.\textsuperscript{21} However, despite the fact that the high-angle boundary length per unit area in the steel aged for 10000 h was the shortest of all of the samples, the $\dot{\gamma}_m$ value for this sample was the second highest in the low-stress region. That is, the high-angle boundary length per unit area, which corresponds to the grain size, did not exhibit such a proportional relationship
in the low-stress region. Therefore, viscous creep cannot be considered as the predominant creep mechanism in the low-stress region. In contrast, the strong correlation between the dispersion state of the $M_23C_6$ particles and the $\dot{\gamma}_m$ value in the low-stress region indicates that creep deformation in this region was also controlled by dislocation motion, as with the high-stress region. However, unlike in the high-stress region, which was strengthened by the martensitic lath structure, the dislocation motion would be prevented by the dispersion strengthening of the $M_23C_6$ particles in the low-stress region. Consequently, although creep deformation is considered to occur by the motion of dislocations in both the stress regions, the creep mechanism is different because the manner of the dislocation motion is considered to be different across the boundary stress of 40 MPa.

4. Conclusions

The creep deformation behavior of Grade P92 ferritic heat-resistant steels which were changed the microstructure by performing long-term aging was investigated with a focus on the transition of the creep deformation mechanism. As a result of this study, the following conclusions were drawn:

1) The stress dependence of the minimum strain rate changed at 40 MPa, even during short-term creep tests, indicating the transition of the creep mechanism at the boundary stress of 40 MPa.

2) Changes in the microstructure occurred because of thermal aging for up to 10000 h at 700°C, such as the precipitation and coarsening of Laves phases, a decrease in the amount of dissolved Mo and W, the precipitation and coarsening of $M_23C_6$ particles, the coarsening of MX particles, and a decrease in the high-angle boundary and martensitic lath boundary lengths per unit area.

3) In the high applied stress region above 40 MPa, the martensitic lath structure had the greatest influence on the minimum strain rate, while in the low applied stress region below 40 MPa, the dispersion state of the $M_23C_6$ particles had the greatest influence. Therefore, it is considered that the difference in the stress dependence of the minimum strain rate reflects the differences in the manner of the dislocation motion and the main obstacles to the dislocation motion in each stress region.

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