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Microstructural evolutions and cyclic softening of 9%Cr martensitic steels

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A B S T R A C T

Detailed TEM and EBSD measurements were carried out to quantify the microstructural evolutions and to identify the physical mechanisms taking place during fatigue and creep-fatigue at 823 K on a P91 martensitic steel. The coarsening of former martensitic laths is shown to be heterogeneous for low applied strains, whereas for higher applied strains and longer holding periods the whole microstructure coarsens. Based on these observations and on a careful study of the stress partition (backstress, isotropic and viscous stress), the softening effect in creep-fatigue is found to be mainly related to the cumulated visco-plastic strain at a given fatigue strain range. The microstructural coarsening taking place during cyclic loadings is shown to increase significantly the minimum creep rate of this steel.

1. Introduction

Martensitic steels of the 9–12%Cr family are widely used in the energy industry and were selected as candidate materials for structural components of future fusion reactors [1,2]. Typical in-service conditions require operating temperatures between 673 and 873 K, which means that the creep behavior of these steels is of primary interest. In addition, some components are anticipated to operate in a cyclic mode, leading to complex time-dependencies of temperature, stress and strain in materials. Therefore, in design procedures, fatigue and creep-fatigue data are required. These steels are known to soften under cyclic loadings [3] due to microstructural instability (lath and precipitates coarsening). Moreover, to meet the need for very long in-service lifetime of components (with very long hold times ~one month) reliable models for cyclic behavior are necessary, since complete tests with such long holding periods cannot, of course, be carried out in laboratory. To make these extrapolations safer and more reliable a detailed understanding of the physical mechanisms responsible for the softening effect is required.

The identification of such mechanisms is firstly based on the study of the macroscopic hysteresis loops through the stress partition between isotropic, kinematic and viscous stress. Secondly, detailed observations carried out at the scale of conventional SEM (EBSD) and TEM enabled the microstructural evolutions to be identified and quantified. The influence of such microstructural instabilities is finally discussed in terms of residual creep resistance.

2. Material and experimental procedures

The material under study is a P91 martensitic steel produced by Usinor. Compositions and heat treatments are given elsewhere [4,5]. Pure fatigue (PF), relaxation-fatigue (RF) and true creep-fatigue (CF) tests (during the holding period, the stress is held constant, which differs from usual relaxation-fatigue tests during which the strain is held constant during the hold time) were carried out at 823 K and controlled on the total strain.

3. Mechanical tests results

Martensitic steels are known to soften under cyclic loadings at high temperature [3,6–12]. This softening effect is illustrated in Fig. 1 where the decrease of the stress range is reported for various applied strain ranges in pure fatigue at 823 K. The softening effect is all the more pronounced and fast when the strain range is high.

However, in creep-fatigue the most relevant comparison between tests is in terms of the cumulated viscoplastic strain. Indeed, as shown in Fig. 2, for a given fatigue strain range ($\Delta\varepsilon_{fat}$), whatever the applied creep strain (ε_{creep}), the softening effect falls along one single 'master curve'. This result highlights the fact that the microscopic phenomena responsible for the softening effect are directly related to the amount of plastic strain applied (glide of dislocations, climb and cross-slip).

Another usual way to study the microscopic phenomena responsible for the macroscopic mechanical behavior is to look at the nature of the stress experienced by the material. Indeed, the macroscopic stress can be divided between the isotropic stress R, the kinematic stress X (or backstress) and the viscous stress σ_v as initially proposed by Cottrel [13] (see Fig. 3). A new stress partition method was developed to obtain such information on highly

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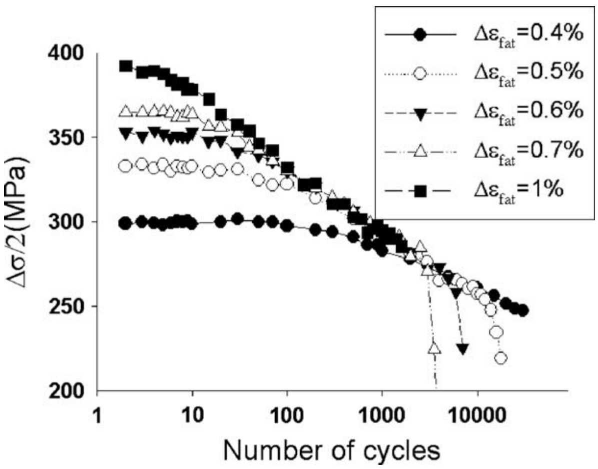

Fig. 1. Variation of the stress range in pure fatigue for various applied strain ranges at 823 K.

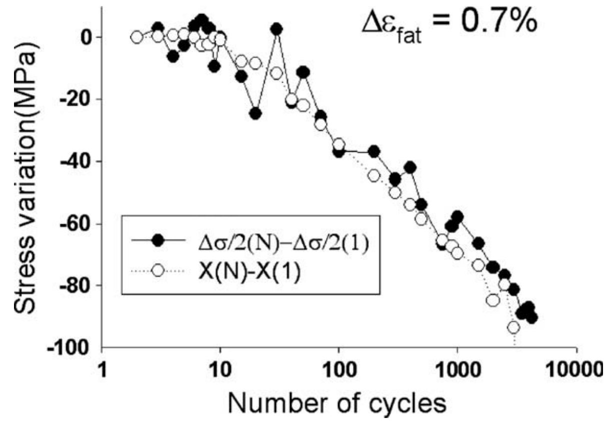

Fig. 4. Correlation between the softening effect and the decrease of the kinematic stress with respect to the first cycle.

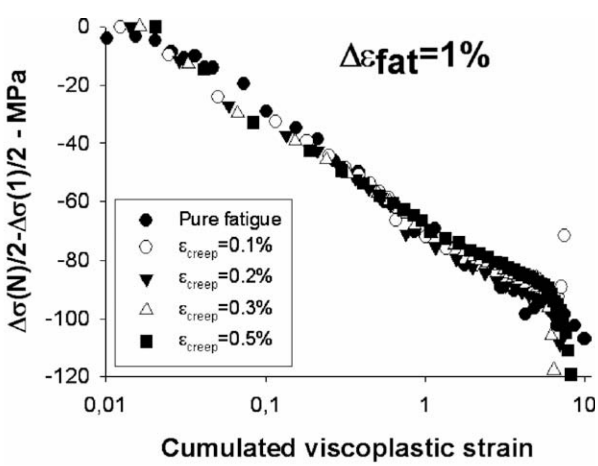

Fig. 2. Variation of the stress range in creep-fatigue at 823 K and $\Delta\varepsilon_{fat}$ = 1% for various applied creep strains.

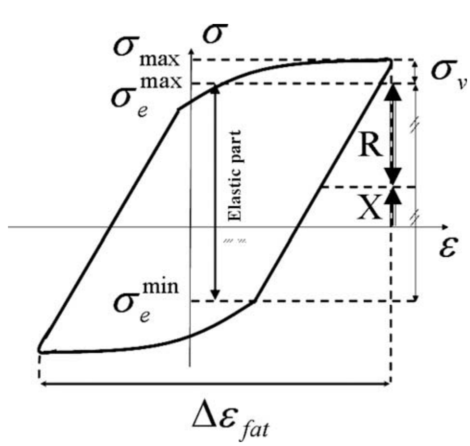

Fig. 3. Scheme of the stress partition obtained from the study of the hysteresis loops in fatigue.

viscous materials [4]. The main results of this study are illustrated in Fig. 4. The softening effect is directly correlated to the decrease of the kinematic stress. This type of stress arises from 'directional and long-range' obstacles to the movement of dislocations. Therefore grain and subgrain boundaries (through a dislocation pile-up mechanism) a and, more generally, all microstructural heterogeneities (microstructure of dislocations, precipitates, etc.) are sources of kinematic hardening. This suggests that, during cycling, the softening effect comes from a disappearance of microstructural heterogeneities.

Finally the study of the macroscopic stress decrease and of the hysteresis loops reveals that the softening effect is directly related to the amount of cumulated (visco)plastic deformation experienced (for a given $\Delta\varepsilon_{fat}$) and to the level of heterogeneity of the microstructure.

4. Observations

In order to confirm the previous results obtained from macroscopic data, detailed TEM and EBSD were carried out.

Fig. 5 presents the evolutions of microstructure observed by TEM. In the as-received condition, several microstructural scales, characteristic of tempered martensite are intricated (former austenitic grain, packet of laths, block of laths, martensitic laths and subgrains) and the finest scale corresponds to subgrains as small as 350 nm with a very high density of dislocations. After pure fatigue at very low applied strain ($\Delta\varepsilon_{fat}$ = 0.3%), some blocks of laths have recovered (very low density of dislocations and almost no subgrain visible) whereas most of the microstructure is unchanged. After creep-fatigue the microstructural recovery is much more homogeneous and all the more pronounced when the applied strain is high. These microstructural evolutions are schematically represented from Fig. 5(e–g). No significant variation (neither coarsening nor precipitation of new phases) of the precipitation state has been observed which was expected for this relatively low temperature and these short testing times (less than three months for each test).

In addition to these qualitative observations of the microstructural recovery, quantitative measurements were carried out using TEM. Distributions of the subgrain sizes in these different states are reported in Fig. 6. In pure fatigue the subgrain coarsening is heterogeneous : the mean subgrain size is only slightly higher than in the as-received condition, but a few very large subgrains (a few microns) were measured. After creep-fatigue the mean subgrain size increases strongly and the whole distribution is shifted to larger diameters. The mean subgrain sizes measured are reported in Table 1 and compared to those measured after creep tests carried out at 823 K and 873 K on various 9–12%Cr steels. It shows that creep-fatigue loadings at 823 K lead to subgrain sizes comparable to those measured after long-term creep at 873 K.

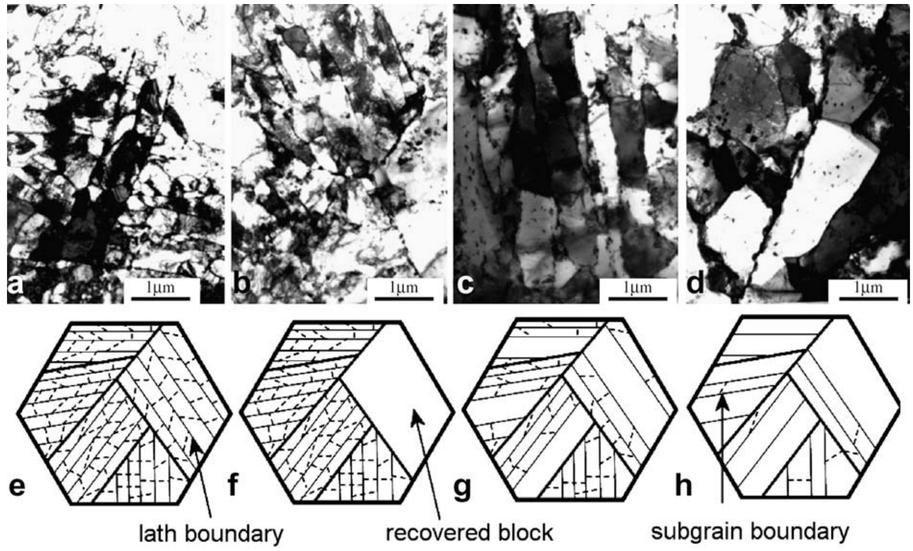

Fig. 5. TEM observations of the microstructure of P91 (a) in the as-received conditions, (b) after low strain pure fatigue ($\Delta\varepsilon_{fat}$ = 0.3%), (c) after low strain creep-fatigue ($\Delta\varepsilon_{fat}$ = 0.4%, ε_{creep} = 0.1%) and (d) after high strain creep-fatigue ($\Delta\varepsilon_{fat}$ = 0.7%, ε_{creep} = 0.5%) and from (e) to (g) corresponding schemes of the microstructure.

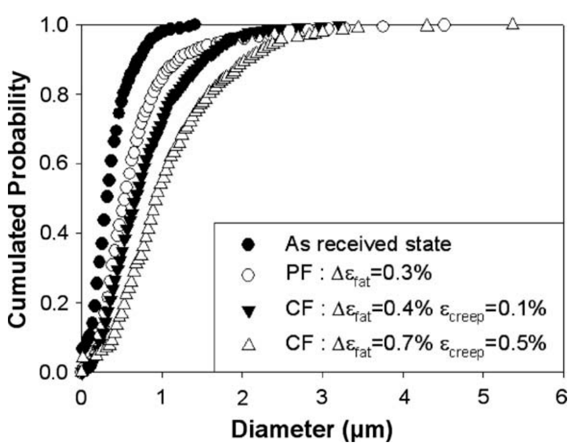

Fig. 6. Distributions of the subgrain diameters in the as-received conditions and after various cyclic tests at 823 K.

EBSD measurements were also carried out on conventional SEM (Fig. 7(a)). Even though the spatial resolution (around 500 nm) is not sufficient to directly measure the evolution at the subgrain scale, an indirect proof of the microstructural coarsening can be obtained when comparing the mean misorientation per block (Fig. 7(b)). Indeed, after RF and CF tests most of the blocks of laths have a mean misorientation lower than 1°, which means that most of the subboundaries initially present inside these blocks have disappeared.

5. Discussion

The previous results highlight the fact that the softening effect measured during cyclic loadings is due to the recovery of the microstructure (decrease of the density of dislocations and coarsening of the subgrains). This confirms the fact that the kinematic stress coming from subgrain and laths boundaries tends to

Table 1
Mean subgrain diameter measured during the present study and taken from the literature after various loadings at high temperatures.

Material	As-received	PF ($\Delta\varepsilon_{fat}$ = 0.3%)	CF ($\Delta\varepsilon_{fat}$ = 0.4%, ε_{creep} = 0.1%)	CF ($\Delta\varepsilon_{fat}$ = 0.7%, ε_{creep} = 0.5%)	Creep at 823 K from [14]	Creep at 873 K from [15–18]
Mean diameter (nm)	372	695	802	1108	450	[730–1800]

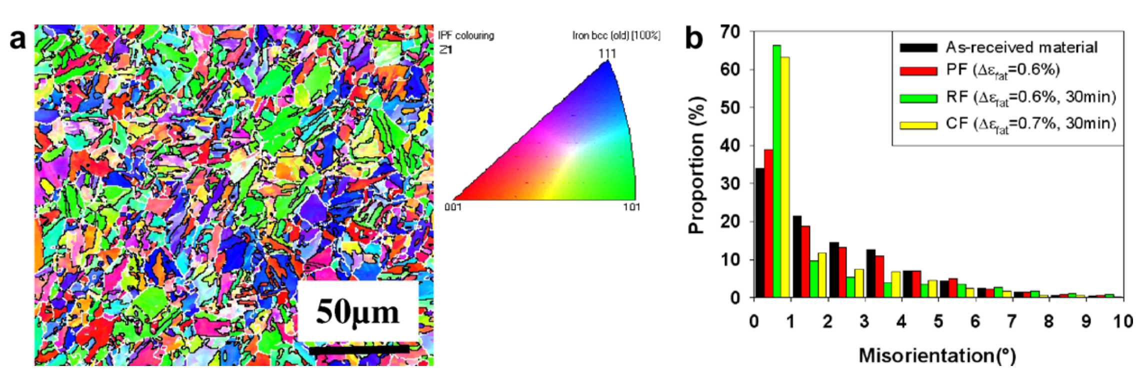

Fig. 7. (a) EBSD map of the as-received microstructure and (b) distribution of the mean misorientations per block of laths after various cyclic loadings at 823 K.

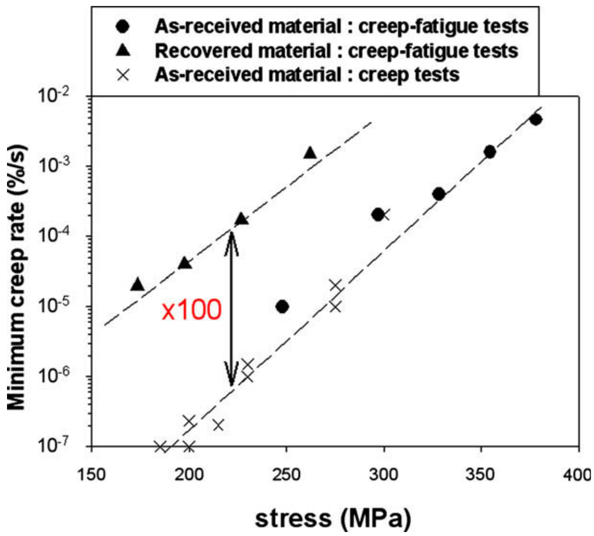

Fig. 8. Comparison between the minimum creep rates measured on the as-received material (creep tests and first cycle of the creep-fatigue tests) and on a recovered material (at mid-life of the creep-fatigue tests).

disappear, whereas the strength coming from precipitates hardening is still constant [4].

A simple Hall-Petch effect can explain qualitatively the lower mechanical strength of a material with a coarsened microstructure. Using the Hall-Petch formulation proposed by Li [19] a first modeling attempt to predict the softening effect gave encouraging results [20,21].

The interaction between creep and fatigue is usually considered, in design procedures, in terms of damage. However, the present tests show that the combination of creep and fatigue deformation significantly modifies the creep behavior (which is the main property of interest for the components under study). Indeed, as shown in Fig. 8, the minimum creep rate measured, during CF tests, at mid-life ($N_f/2$), can be 100 times faster than for the as-received material. Which means that a material that recovered due to cyclic loadings has a poorer creep strength [22]. Such variation of the creep behavior should be taken into account in design procedures.

Moreover, the microstructures observed after creep-fatigue present a clear similarity with those observed after long-term creep. Indeed, in both cases, neither subgrains nor laths can be observed anymore. In the case of long-term creep, an additional evolution of the microstructure takes place: the precipitates coarsen and new phases appear. Therefore, sequential tests with a prior creep-fatigue loading, followed by a creep test, could be used as 'accelerated' creep tests (creep-fatigue preloading lasts only a few days). Such tests would enable to quantify the acceleration of the creep rate due to the disappearance of the dislocations structure. A creep-fatigue preloading followed by an ageing at higher temperature (which would lead to the precipitation of new phases and to a coarsening of precipitates) would enable the mechanical properties of 9–12%Cr steels after long-term in-service exposure (mainly under creep conditions) to be tested and used for design codes.

6. Conclusions

The study of the macroscopic behavior (hysteresis loops and stress variations) combined to detailed microscopic observations carried out on samples previously subjected to fatigue and creep-fatigue tests at 823 K enabled the following conclusions to be drawn:

- The softening effect observed on martensitic steels is directly correlated to the applied (visco)plastic strain.
- This softening behavior is due to the disappearance of most of the microstructural subboundaries characteristics of the tempered martensite and to the decrease of the density of dislocations.
- This microstructural coarsening is heterogeneous for low applied strain and all the more homogeneous and pronounced when the applied plastic strain is high.
- The quantification of this coarsening showed that creep-fatigue loadings lead to similar microstructural size than creep loadings, but this coarsening is much faster in creep-fatigue (a few weeks) than in creep (a few months/years).
- The recovered material has a much poorer creep strength.

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