The stress amplitude of serrations first increases with decreasing strain rate and then gradually saturates. The matrix carbon concentration affects the stress amplitude and the tendency to saturation.

The effects of strain rate and carbon concentration on the dynamic strain aging of cold rolled Ni-based alloy in high temperature water

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Abstract

The effect of strain rate on dynamic strain aging of cold-rolled Ni-based alloy was investigated. With decreasing strain rate, the stress amplitude of serrations first increased and then saturated. Compared with the solution-annealed condition, the thermally-treated condition produced smaller stress amplitudes that saturated at a lower strain rate. Observations are consistent with a mechanism in which the locking strength of solute atmospheres first increases with increasing solute atom arrival at dislocations and gradually saturates as solute reaches a critical level.

Keywords: cold rolling; slow strain rate tensile test; nickel alloys; dynamic strain aging; Portevin – Le Chetelier effect

Serrated plastic flow (dynamic strain aging (DSA) or Portevin-Le Chatelier (PLC)) is observed in many alloys during deformation in specific temperature and strain rate ranges. It is widely believed that it arises from the interaction between diffusing solute atoms and mobile dislocations that are temporarily impeded [1, 2]. Much previous work has been done to investigate the influence of different variables on DSA. Related work [3-12] on different alloys show that increasing strain rate results in decreasing amplitude of the stress drop. The lowest strain rate applied in those works was 10^{-7} s^{-1} . Because many important phenomena, such as stress corrosion cracking, occur at strain rates below this level, it is of interest to understand DSA behavior at lower strain rates. The alloy of interest in this work is Alloy 690 which is widely used in nuclear power plant components exposed to high temperatures and low strains. The typical creep rate of this alloy in simulated pressurized water reactor primary water environment is in the range of 10^{-10} to 10^{-9} s^{-1} [13]. The effect of carbon concentration was also studied by testing samples subjected to different thermal treatments. To date, very limited work has been done to study the DSA of this alloy [9, 14, 15].

The chemical composition of Alloy 690 used in this work is 57.6 wt.% Ni, 32.7% Cr, 8.64% Fe, 0.25% Mn, 0.315% Al, 0.08% Si and 0.02% C. The alloy was received as a forged bar with a diameter of 185 mm. It was solution annealed (SA) at 1100 °C for 1 h and then water quenched. In order to change the carbon concentration in matrix, some solution annealed samples were thermally treated (SA+TT) at 700 °C for 17 h and cooled in air. Both SA and SA+TT samples were

then cold rolled (CR) to 20% thickness reduction, resulting in a sheet of approximately 8 mm in thickness. The cold rolled sheet was machined into round tensile bars with the sample axis in the rolling direction. The gage section of the tensile bar is 20 mm in length and 2 mm in diameter. Samples were mechanically abraded up to 4000 grit and then electropolished for 30 seconds at 30 V in a solution of 10% (volume fraction) perchloric acid in methanol. Some coupons were also prepared for carbide analysis using the same procedure. Grain boundary carbides were characterized with scanning electron microscope (SEM) on a FEI Helios Nanolab 650. Tensile bars were strained to failure in 360 °C high purity water containing 1.62 ppm hydrogen (by weight) which is approximately at the electrochemical potential of the Ni/NiO phase boundary. The inlet resistivity was above 18 MΩ·cm and the outlet resistivity was above 5 MΩ·cm. A recirculating water loop equipped with a 4 L stainless steel autoclave was used to maintain the required water environment. Samples were strained with a 5K servo via a crosshead that was capable of loading a maximum of 4 tensile bars.

Figure 1 shows the SEM images of these two samples. The SACR sample shows no significant grain boundary carbide precipitation while SA+TTCR sample has dense semi-continuous carbides. The remaining C concentration in SA+TTCR sample was estimated using the computer code DEplete690 [16]. The code contains a thermodynamic and kinetic analysis of grain boundary carbide precipitation in alloy 690 as a function of alloy content and time at temperature and has been benchmarked against experimental measurement [16]. From the calculation, the C

concentration in the matrix decreased from 0.94 at.% to 0.78 at.% after the thermal treatment at 700 °C for 17 h.

After the desired water environment stabilized, the samples were first loaded to just below the yield point at a rate of $1.24 \times 10^{-5} \text{ s}^{-1}$ and then strained to failure at various extension rates. Four nominal strain rates were used: $2.5 \times 10^{-7} \text{ s}^{-1}$, $5 \times 10^{-8} \text{ s}^{-1}$, $2.5 \times 10^{-8} \text{ s}^{-1}$ and $1 \times 10^{-8} \text{ s}^{-1}$. The samples were designated by the thermal mechanical treatment and applied strain rate. For example, the solution annealed and cold rolled sample strained at $2.5 \times 10^{-7} \text{ s}^{-1}$, $5 \times 10^{-8} \text{ s}^{-1}$, $2.5 \times 10^{-8} \text{ s}^{-1}$ and $1 \times 10^{-8} \text{ s}^{-1}$ was designated as SACR-0, SACR-1, SACR-2 and SACR-3, respectively.

Figures 2a and b show the stress-strain curves for SACR and SA+TTTCR samples at the four different strain rates. For better view, the curves on each graph were separated and the two upper curves share the right y axis. All the y axes have the same scale spacing. The rising part of the serrations is elastic and the apparent modulus is unchanged with strain rate. Serrations occurred shortly after yielding, and by their shape, fall into the definition of type C which is due to dislocation unlocking [1]. R. W. Hayes and W. C. Hayes [7] reported that the activation energy of DSA for a nickel-based alloy (Waspalloy) in the temperature range of 204-649 °C is consistent with a carbon atmosphere aging mechanism. The work of Hänninen et. al [14] also suggests that the serrated plastic flows of alloy 600 and 690 at temperature of 100-600 °C result from carbon-dislocation interaction. So it is believed that the serrations in alloy 690 at 360 °C are due to carbon atmospheres formed at dislocations.

Serrations occurred in different plastic strain ranges for different samples. To compare different samples, only the serrations within the same plastic strain range (0.4%-4%) were selected to calculate the average stress amplitudes. The plastic strain was calculated by subtracting elastic strain and compliance from the total strain. All the serrations within this plastic strain range for each sample were well developed and unaffected by the failure of other samples loaded at the same time. The arrows in Figure 1a and b denote the starting and ending serrations used for the calculation of average stress amplitudes of serration. The change in the average stress amplitudes as a function of strain rate is shown in Figure 2c. For the same strain rate, SA+TTCR sample has smaller average stress amplitudes of serration than SACR sample. The average stress amplitude increased with decreasing strain rate from $2.5 \times 10^{-7} \text{ s}^{-1}$ to $2.5 \times 10^{-8} \text{ s}^{-1}$ for SA+TTCR samples and saturated as the strain rate was further decreased to $1 \times 10^{-8} \text{ s}^{-1}$. For SACR samples, the average stress amplitude increased from $2.5 \times 10^{-7} \text{ s}^{-1}$ to $5 \times 10^{-8} \text{ s}^{-1}$ and saturated with some fluctuations as the strain rate was further decreased. It would be ideal to produce more data to consolidate this finding if additional samples were available. Nevertheless the results still clearly suggest a trend of stress amplitude plateau towards lower strain rate for both conditions. The stress amplitude saturates at higher strain rate on SACR than on SA+TTCR sample, suggesting that such saturation effect is dependent on the microstructure of the material. This is the first reported observation of a saturation effect in DSA.

Figure 3 shows the change in stress amplitude as a function of plastic strain for the

case of SACR (Fig. 3a) and SA+TTTCR (Fig. 3b). The points at which necking occurred are marked with arrows. To decrease the noise level of the stress amplitude of serrations, a 5-point moving average was applied on the raw data. For both conditions, the stress amplitude increased with plastic strain. Similar results have been reported in Al alloys [5, 17]. In addition to total plastic strain, prior cold work has also been found to increase the amplitude of the stress drop in Al alloys and Alloy 800H [4, 6, 9]. It is generally accepted that the diffusion of solute is enhanced by increasing vacancy concentration due to plastic strain, and the amplitude of stress drop is proportional to the number of solute atoms arriving at the dislocation sites [5]. The serration amplitude did not change abruptly once the sample necked although the strain distribution became more complicated. The SACR samples basically showed the same trends although there are some fluctuations (Figure 3a).

Figure 3b shows that the stress amplitude increased with decreasing strain rate from $2.5 \times 10^{-7} \text{ s}^{-1}$ to $2.5 \times 10^{-8} \text{ s}^{-1}$ for the SA+TTTCR condition. Similar results have been reported on Fe + 3.0 at.% Ni [3], 5086 Al alloy [4], Al-5 wt.% Mg alloy [5], Al-Mg5-Mn0.8 alloy [6], Al-5at.%Mg alloy [8], alloy 600, Waspalloy [7] and alloy 690 [9]. Cuddy and Leslie [3] proposed that increasing the loading rate would decrease the aging time and thus the locking strength of solute atmospheres. Korbel and Dybiec [6] reported similar results on Al-Mg5-Mn0.8 alloy and tried to explain the effect of strain rate by considering the relaxation on internal stress on the deformation band. However, they ignored the change in locking strength of solute atmosphere under different strain rates. Robinson and Shaw [2] pointed out in their

review that increased strain rate would lessen the waiting time of mobile dislocations temporarily arrested by local obstacles (forest dislocations or precipitates) and result in a decreased strength of the forest/mobile dislocation junction. For SACR samples (Figure 3a), a similar effect of strain rate was observed from $2.5 \times 10^{-7} \text{ s}^{-1}$ to $5 \times 10^{-8} \text{ s}^{-1}$. However, as the strain rate was further decreased from $2.5 \times 10^{-8} \text{ s}^{-1}$ to $1 \times 10^{-8} \text{ s}^{-1}$ for TTCR sample (Figure 3b) and from $5 \times 10^{-8} \text{ s}^{-1}$ to $1 \times 10^{-8} \text{ s}^{-1}$ for SACR sample (Figure 3a), the stress amplitudes changed slightly. Such results are very consistent with Figure 2.c. It is possible that when the strain rate is low enough, the number of carbon atoms in solution diffusing to the dislocation site would saturate and the locking strength of the solute atmosphere would also saturate.

According to the work of Cottrell and Bilby [18], the accumulation rate of solute at dislocation site is dependent on the average concentration and diffusivity of solute in solution. Assuming that a critical solute concentration at a dislocation site is required for the locking strength to saturate, it follows that a higher average concentration of solute in solution will result in a shorter time to saturation of locking strength. The SACR samples have higher C in solution than the SA+TTCR samples which underwent significant carbide precipitation at grain boundaries during the thermal treatment (Figure 1). The larger amount of C in solution in the SACR sample explains the higher stress amplitude compared to the SA+TTCR sample. It also explains why the stress amplitude started to saturate in the SACR sample at a higher strain rate than in the SA+TTCR sample (Figure 2.c). It should be noted that the samples discussed here were cold rolled and the diffusivity of C in those samples

should be faster than in the non-cold rolled counterparts. So the saturation of DSA may not be observed in the non-cold rolled samples strained at the same rates.

The effect of strain rate on the stress amplitude of serrations can be understood from Figure 4. In this figure, the average stress amplitude of serration from Fig. 2c, is plotted against the average waiting time (the period when the mobile dislocations are temporarily arrested at local obstacles). The waiting time was calculated by dividing the average strain between serrations by the strain rate. The dashed lines indicate the total anchoring strength which is the sum of the locking strength of solute atmosphere on dislocations temporarily impeded by the local obstacle, and the arresting strength of the obstacle. The straight lines represent the applied stresses at different strain rates with reference to the minimum stress level of serrations. Note that the elastic modulus is slightly different between these two samples. When the applied stress surpasses the total anchoring strength, serration occurs. The total anchoring strength increases with waiting time and then saturates as the solute atoms accumulate at the dislocation site and reach the critical level. A lower strain rate allows for longer waiting time and results in a higher stress amplitude until saturation occurs. The SACR sample has a higher anchoring strength and reaches saturation sooner than the SA+TTTCR sample due to its higher carbon content in solution. So the SACR sample has higher stress amplitude of serration compared to the SA+TTTCR sample at the same strain rate.

In conclusion, the stress amplitude of serration increased with plastic strain due to enhanced diffusivity of carbon. For the same strain rate, the SACR sample has a

higher stress amplitude compared to the SA+TTCR sample due to its higher carbon content. The stress amplitude of serration increases with decreasing strain rate until saturation occurs. The stress amplitude starts to saturate in the SACR sample at a higher strain rate than in the SA+TTCR sample. The saturation of DSA was observed experimentally for the first time. The observations are consistent with a mechanism in which the locking strength of solute atmosphere first increases with increasing number of solute atoms arriving at the dislocation site, and gradually saturates as the solute concentration approaches a critical value. Consequently, the stress amplitude of serration first increases with decreasing strain rate, which results in an increased waiting time, and then levels off when the strain rate is low enough to for saturation to occur.

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Figure captions:

Figure 1. SEM images of (a) SACR and (b) SA+TTTCR samples.

Figure 2. Serrations in stress-strain curves of (a) solution annealed and cold rolled and (b) solution annealed+thermally treated and cold rolled Alloy 690 strained at different rates in 360 °C water (the vertical arrows indicate the starting and ending serrations used for the calculation of average stress amplitudes, and the horizontal arrows indicate the stress scale for the plot); (c) the changes of average stress amplitudes of serration (in the plastic strain range of 0.4%-4%) with strain rate.

Figure 3. Changes of stress amplitudes of serrations of (a) SACR and (b) SA+TTTCR samples over plastic strain.

Figure 4. Schematic of the change of stress amplitude of serrations with waiting time for different strain rates.

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