The Role of Cold Working in the Creep Deformation of Nb Stabilized Stainless Steel

I. Creep Results and Microstructural Evolution

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ABSTRACT

The effect of cold working on the creep strength and microstructural evolution of an AISI 347 stainless steel during creep was studied between 625 and 825°C. The relationship between the minimum creep rate, the stress and the temperature was obtained by best fitting of the experimental data. The microstructural parameters (average dimension of the particles, number of precipitates per unit volume, dislocation density) were measured in samples crept up to rupture as well as after interrupted tests; the Orowan stress obtained from microstructural data was related to the rupture dislocation density. The effect of prestraining on the deformation-time curve was found to be different from the one observed in other stainless steels. These results and the microstructural observations suggested that the precipitation of fine NbC played an important role in reducing the kinetics of the recovery processes.

INTRODUCTION

The effect of prestrain on the creep properties of several materials was extensively studied /1-5/. Cold working introduces a high initial dislocation density, which in austenitic stainless steels generally gives rise to a substantial increase in creep strength and rupture life. The evolution of dislocation density during creep

of a coarse grained material /6/ showed an initial strong increase in the free dislocation content (free dislocations being the fraction of dislocation population not arranged in subgrains) followed by the re-arrangement of the dislocations in networks, and subsequently in subgrain boundaries. Extensive research has been carried out in order to understand the creep mechanisms in class M materials (pure metals and single phase alloys), in which creep deformation is recovery-controlled /7,8/. Recovery creep was explained in terms of the interaction of strain hardening and recovery processes, leading to the formation of a three-dimensional network of dislocations. The high creep strength shown in cold worked AISI 304 /5/ or Fe-21Cr-32Ni /4/, for example, was attributed mainly to the entanglement of dislocations into a three-dimensional network of dislocations (Taylor hardening); nevertheless, the precipitation of fine $M_{23}C_6$ on dislocations was observed for long exposure times in not prestrained as well as in cold worked materials /9,10/. When precipitation occurred in these simple austenitic stainless steels, secondary creep was thought to be controlled by particle-dislocation interaction, while primary creep was associated with different phenomena, such as the formation of planar arrays of dislocation, the viscous drag of Cr and C solute atoms and piling up of the line defects /9/.

The study of the creep response becomes even more complex in the Nb or Ti bearing stainless steels. These

steels are stabilized against Cr carbide precipitation (sensitization) by small additions of Nb or Ti, so that when tested in the solution-treated state, an extensive precipitation of fine Nb or Ti carbides nucleated on dislocations is invariably observed [3,11-14]. On this basis, the improved creep behaviour of the solution-treated and stabilized material was attributed to the particle strengthening mechanisms [3,11,15-19]; moreover, when the material was prestrained before aging or creep, a significant increase both in the precipitation kinetics as well as in creep strength was observed [3,11,13,19].

This study aims to evaluate the effect of a prestraining on the creep response and on the microstructural evolution of the AISI 347 stainless steel.

MATERIAL AND EXPERIMENTAL

The material used in this study was AISI 347. This steel, supplied in the form of extruded tubes in a solution-treated condition (1120°C-45 min), had the following chemical composition (weight %): 17.3 Cr, 10.4 Ni, 0.048 C, 0.52 Nb, < 0.01 Ta, Fe balance.

Creep specimens with a gauge length of 50 mm were machined from the tube and then cold worked in tension up to 5, 10 or 15% plastic deformation; before creep the samples were remachined up to the final diameter of 5 mm.

The constant-load creep tests were carried out in air at a temperature between 625 and 825°C under stresses of 50, 127, 157 and 196 MPa; some of the tests carried out at 650°C and 157 MPa were interrupted after 24, 120 or 600 h and cooled under load.

A detailed characterization of the microstructure was performed by TEM; slices cut with a diamond saw at 45° to the longitudinal direction of the crept samples were ground to a thickness of 200 µm, and then electro-polished to 50 µm thickness with a 10% perchloric acid in an ethylene glycol solution at 12°C and 25V. Thin foils were subsequently obtained by double jet in the same solution.

The number of particles per unit volume N_v was estimated by an image analysis system; the dislocation density ρ was measured on TEM micrographs by the

intercept method. The Orowan stress was estimated from the equation [20,21/

$$\sigma_{OR} = 3.06 \frac{G \cdot b}{2\pi \lambda \sqrt{1-\nu}} \ln \left(\frac{d_s}{b} \right) \quad (1)$$

where G is the shear modulus, b is the Burgers' vector, ν is the Poisson ratio, λ is the mean interparticle spacing calculated from N_v and the average dimension of the particles d [20/ and $d_s = d(2/3)^{0.5}$.

RESULTS

Creep Results

The creep curves shown in Fig. 1 illustrate an example of the effect of cold working (c.w.) on creep deformation observed between 625 and 725°C. The prestraining induced a substantial reduction in the minimum creep rate ($\dot{\epsilon}_m$), an increase in the time to rupture (t_R) (Figs. 2-3 and Table 1) and a dramatic decrease in the rupture ductility.

The minimum creep rate was related to stress and temperature by the equation:

$$\dot{\epsilon}_m = A \sigma^n e^{-\frac{Q}{RT}} \quad (2)$$

where Q is the apparent activation energy for creep, R the gas constant, T the absolute temperature and A and n parameters depending on the material. The apparent activation energy for creep was found to show a weak dependence on cold working amount [22/], the average value being 434 kJ/mol. This is fully comparable with the value of 398-464 kJ/mol obtained in other austenitic stainless steels strengthened by NbC particles [11/]. The stress exponent n was 8.8, 6.8 and 4.9 for as received (a.r.), 5% cold worked (c.w.) and 15% c.w. materials, respectively; a similar decrease in n as the amount of cold work increased was also observed for the 20Cr-25Ni-Nb stainless steel [3/].

The minimum creep rate and the time to fracture were related by the Dobes – Milička equation [23/:

$$\log \left(\frac{t_R}{\epsilon_R} \right) + m \log \dot{\epsilon}_m = C \quad (3)$$

Fig. 1: Creep-rupture curves at 675°C-196 MPa for the as-received, 5 and 15% cold worked samples.

Fig. 2: Effect of cold working on time to fracture.

Fig. 3: Effect of cold working on the minimum creep rate.

Fig. 4: Dependence of ratio of time to fracture to total creep deformation on minimum creep rate.

Fig. 5: Effect of cold working on the shape of the creep curve at 825°C.

where ϵ_r is the deformation at rupture, and C and m (m usually close to 1) are material parameters. The computed value of m is 1.013 (Fig. 4); the proportionality of the product of minimum creep rate and time to fracture to the total creep deformation could be considered, as shown by Dobes and Milička /23/, a confirmation of the deformation dependence of rupture processes.

The trend of the experimental creep curves recorded at 825°C and 50 MPa is reported in Fig. 5: the 15% c.w. creep curve after 20 h showed an increase of creep rate that led to a shorter time to rupture. Moreover, the relatively large strain to fracture (about 40%) is practically unaffected by c.w.

Table 1
Minimum creep rate and time to rupture obtained for as-received, 5 and 15% c.w. samples

c.w. %	T [°C]	σ [MPa]	t_R [h]	$\dot{\epsilon}_m$ [10^{-6} h^{-1}]
0	625	196	1539	18
	650	196	327	100
	675	196	126	309
	700	127	411	35.2
	725	127	120	200
	825	50	123	568
5	625	196	1714	7.56
	650	196	675	20
	650	157	1922	3.9
	650	127	4200	1.7
	675	196	159	142
	700	196	58	425
	700	127	535	20.3
	725	127	128	133
15	825	50	180	465
	625	196	2597	2.21
	650	196	794	12.3
	650	196	777	9.5
	650	196	699	9.1
	650	157	1670	4.0
	650	127	5107	1.3
	675	196	298	57.6
	700	196	78	236
	700	127	604	26.3
	725	127	260	112
	825	50	98	297

Microstructure

Fig. 6 shows the microstructure of the as-received material obtained directly from the tube: large Nb-rich primary particles, associated with dislocation tangles, were dispersed in the matrix. After prestraining, the dislocation density dramatically increased ($\rho > 10^{14} \text{ m}^{-2}$).

The microstructure after creep rupture at 196 MPa-650°C is shown in Fig. 7: an extremely high dislocation density still persisted after $t > 1000$ h, and recovery was far from complete. At a higher magnification, there was evidence of a fine precipitation of NbC particles decorating line defects.

The interrupted tests were used to investigate the microstructural evolution during creep exposure at 650°C (Figs. 8-10 and Table 2). Between 24 and 120 h,

Fig. 6: Microstructure of the as-received material.

Fig. 7: Microstructure of the crept sample at 650°C-196 MPa: a) as received; b) c.w. 5%; c) c.w. 15%.

Fig. 8: Microstructure of the as received material during creep at 650°C-157 MPa; a) 24 h; b) 120 h; c) 600 h.

the dislocation density progressively increased, reaching a maximum, and at the same time fine particles began to nucleate on dislocations (Figs. 8-10 a and b). After 120 h, the dislocation density dropped, and in the as-received as well as in 5% c.w. samples growth of the intragranular particles was observed. In the 15% c.w. material the dimension of the NbC did not increase significantly between 120 and 600 h; it should be pointed out that in the highly prestrained samples the

number of particles per unit volume shown in Table 2 was probably underestimated, due to the difficulty in unambiguously identifying the finest particles (size < 10 nm)

Fig. 11 shows the microstructure after rupture at 825°C; a very well defined substructure appeared, the dislocation density being very low both in a.r. and in 15% c.w. materials. The dimension of secondary intragranular particles was finer in the 15% prestrained

Fig. 9: Microstructure of the 5% c.w. material during creep at 650°C-157 MPa; a) 24 h; b) 120 h; c) 600 h.

Fig. 10: Microstructure of the 15% c.w. material during creep at 650°C-157 MPa; a) 24 h; b) 120 h; c) rupture.

sample, and precipitation was slightly more extensive than in the as-received and crept material. The high test temperature and the high initial dislocation density in 15% c.w. steel made the recovery processes very fast, producing a substructure that was roughly equivalent to the as-received one; this softening resulted in a gradual loss of creep strength and in the early onset of

accelerated creep. On the other hand, the higher dislocation density in the subgrain interior in the 5% c.w. material after rupture gave evidence of a reduced driving force for annealing. The high creep strength of the 5% c.w. observed at this temperature could consequently be explained in terms of microstructural stability, i.e., slower kinetics of recovery.

Table 2
Microstructural parameters obtained from interrupted tests at 650°C-157 MPa.

c.w.	time [h]	ρ [10^{14}m^{-2}]	N_v [10^{21}m^{-3}]
0	24	2.04	>0.04
	120	2.62	0.5
	600	1.00	0.3
5	24	3.52	>0.1
	120	4.58	0.6
	600	2.52	0.8
15	24	4.27	>0.4
	120	5.43	>0.9
	600	4.46	>1.0

Fig. 11: Microstructure of the sample crept at 825°C-50 MPa; a) a.r.; b) 5% c.w.; c) 15% c.w.

DISCUSSION

Both recovery and particle growth were observed to be relatively slow in the highly prestrained material between 625 and 725°C. Kesternich /19/ showed a similar effect in a 15% c.w. DIN 1.4970 steel. The decrease in TiC dimension as c.w. increased was attributed by the author to an increase in the number of nucleation sites and to a reduction in dislocation mobility. In fact, the dislocations, dragging the Cottrell atmosphere, generated a local supersaturation of solute atoms at the point where the dislocations interacted,

leading to the nucleation of particles; a significant increase in the particle dimension was observed only as long as the growth was assisted by the mobile dislocations (recovery-controlled growth). A high initial dislocation density was produced by prestraining enhanced nucleation of fine particles that pinned the dislocations, reducing the kinetics of recovery. On the other hand, when the initial dislocation density was low, the effectiveness of particle nucleation was reduced, and as a result the particle pinning effect was also reduced. In this sense nucleation was promoted in the prestrained samples, while recovery-controlled

growth /19/ was promoted in the as-received material.

In order to relate the dislocation density to the characteristics of particle distribution, the Orowan stress obtained at rupture at 625°C-196 MPa, 650°C-196 MPa and 725°C-127 MPa was reported as a function of $Gb(\rho)^{0.5}$ (Fig. 12). The correlation between the two parameters at 625 and 650°C was sought in the form

$$\sigma_{OR} = \alpha Gb\sqrt{\rho} \quad (4)$$

The calculated value of the constant α was 0.95. Equation 4 is formally similar to the relationship proposed by Ecob /16/ to relate the Orowan stress to the "irrecoverable dislocation density" ρ_0 (i.e., a threshold dislocation content that depends on the particle distribution, and is equivalent to the annealed in dislocation density). The value of ρ_0 is such that the average dislocation link length is approximately equal to λ /17/; according to Ecob, when the applied stress is lower or close to the Orowan stress, the free dislocation density during the secondary and tertiary stage is constant and equal to ρ_0 . Senior /15/ reported a similar behaviour in an AISI 347 tested at 650°C. The data obtained in the present study at 650°C from the interrupted tests were consistent with this theory; during primary creep the recovery processes reduced

the dislocation content, increasing the mean dislocation link length $L \propto (\rho)^{-0.5}$ in the network /24/. When L becomes equal to the interparticle spacing λ , the recovery processes are obstructed by the particle-dislocation interaction and the dislocation content remains almost constant up to rupture; then, with $\lambda = L \propto (\rho)^{-0.5}$, the following equation is obtained /16/:

$$\sigma_{OR} \propto \frac{Gb}{\lambda} \approx \alpha Gb\sqrt{\rho} \quad (5)$$

where α is a geometrical factor close to 1. Nevertheless, at 725°C the experimental data lie well above the line that represents Eqns. 4-5; this result suggests that at high temperature the microstructural evolution was somewhat different from that at 625 and 650°C. This behaviour was much more evident at 825°C, where the distribution and dimension of the fine NbC particles were found similar in the a.r. and 15% c.w. materials (N_v was indeed slightly larger in the prestrained sample), while the subgrain interior was practically dislocation-free. At a high temperature, recovery of highly prestrained materials is very fast, and combination of the different processes of recovery of dislocation substructures-nucleation and growth of the NbC results in very well defined substructures and relatively large particles.

The lower minimum creep rate, shown by the 15% prestrained material with respect to the a.r. and 5% c.w. ones between 625 and 725°C, could be attributed mainly to Taylor hardening /5/ and/or to the extensive precipitation of fine particles (particle strengthening) /11,22/. Nevertheless, the creep behaviour of this material was the result of a complex balance of different and sometimes simultaneous and interlaying mechanisms:

- i) the introduction of a high dislocation density due to prestraining, which results in the formation of a relatively stable network of dislocations (Taylor hardening);
- ii) the recovery processes due to the high temperature, and the subsequent formation of substructures and reduction of the free dislocation density;
- iii) the precipitation of fine NbC, which nucleated on dislocations and obstructed the mobility of

Fig. 12: Relationship between the Orowan stress and the dislocation density measured at fracture; the data obtained at 725°C were not used in the calculation of the α constant.

- the line defects, reducing the kinetics of recovery;
- iv) the drop of the amount of elements in solid solution (mainly Nb and C) that occurred in parallel with precipitation, resulting in the reduction of solid solution strengthening;
 - v) the growth of the fine particles, that led to an increase in particle spacing and to a reduction of the particle strengthening effect.

Creep Deformation in the As-Received (Solution-Treated) Material

Fig. 13 shows an interesting comparison between the a.r. creep response observed in this study and the creep curves reported by Senior /15/ under similar test conditions for an AISI 347 after solution treatment and 750°C x 200 h aging. After an extremely fast (typically 10 h at 650°C) decrease in the creep rate, the solution-treated (a.r.) and crept material showed a pseudo-tertiary response (i.e., a continuous, even if very slow, increase in the creep rate). During the very early stage of creep deformation ($t < 50$ h), a relatively high content of Nb and C was retained in solid solution; on the other hand, the results of measurements of dislocation density suggested that ρ increased, reaching a maximum between 24 and 120 h. On this basis, the onset of the pseudo-tertiary creep could perhaps be

attributed to a class A (solid solution) creep behaviour (both dislocation density and strain rate increased with time). As deformation (and time) proceeded, precipitation occurred (Fig. 8) and the solid solution strengthening lost its effectiveness; moreover, the dislocation density dropped (Table 2) and creep became recovery-controlled (class M). The slow increase in creep rate after 100 h, in this case, could be attributed to different phenomena (i.e., coarsening of the particles, reduction in the fraction of mobile dislocation density). The comparison in Fig. 13, on the other hand, shows that aging considerably increased the creep strength of a similar material; the creep curves obtained by Senior /15/ had a relatively long normal primary creep stage, and the creep rate attained a minimum value that is lower than in the a.r. and crept samples. This result could be expected due to the strengthening effect of the fine NbC particles in the 750°C aged steel (average particle dimension 4.5 nm), which is more effective than in the solution-treated and crept samples of this study ($d = 50$ nm at $t = 600$ h).

Creep Deformation in Cold Worked Material

Increasing the cold working amount changed the shape of the creep curve, and a normal long primary creep stage appeared. This result is in direct contrast with previous observations on the creep response of

Fig. 13: Comparison between the creep response of the as received material and the creep curves obtained by Senior /15/ under similar test conditions from an AISI 347 aged steel.

cold worked materials /25/, including stainless steel like the AISI 304 /26/. The pseudo-tertiary creep, typical of prestrained materials, was generally attributed to rearrangement of the dislocations introduced by cold working; on the other hand, the results of the present study suggest that the observed long primary creep corresponded to a sharp decrease in dislocation density, the shape of the 15% c.w. creep curves being similar to the one reported by Senior for the aged material. The similarities in the shape of the creep curves in solution-treated-c.w. and in solution-treated-aged materials seem to indicate that deformation is controlled by the same mechanism (presumably particle strengthening). Moreover, even if the free dislocation density decreased during creep, a fully recovered substructure was obtained only at a very high temperature (825°C). On the other hand, in room /26-29/ as well as in high /30/ temperature prestrained materials, where precipitation of fine MX is not expected, as in AISI 304 stainless steel, the Taylor hardening is the main strengthening mechanism, and the shape of the creep curve is exclusively controlled by evolution (i.e., re-arrangement) of dislocation in well defined substructures, leading to the onset of pseudo-tertiary creep.

CONCLUSIONS

Cold working significantly improved creep strength (time to rupture) but decreased rupture ductility; full recovery of microstructure, that led to a dramatic decrease in creep strength, was observed in the cold worked material only at the highest test temperature (825°C).

This study attempted to establish the following qualitative interpretation of the creep behaviour of cold worked material between 625 and 725°C: after a sharp decrease in creep rate, which corresponded to an increase in dislocation density content, a long primary transient stage was accompanied by a recovery-controlled decay of free dislocation density and by the precipitation of NbC. The high initial dislocation density, in fact, promoted the nucleation of very fine (<10 nm) NbC, that obstructed the slip of dislocations, reducing the re-arrangement in substructures. When

the average dislocation link length was approximately equal to the interparticle spacing, the dislocation density attained a constant value, and deformation became climb-controlled, the fine particles being the most effective strengthening mechanism.

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