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EFFECT OF HELIUM
ON CREEP OF TYPE 316 STAINLESS STEEL
AT 700°C

AEC Research and Development Report

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AT 700°C

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ABSTRACT

Small helium-injected samples of Type 316 stainless steel were creep tested in vacuum, under constant load conditions, at 700°C, with an initial stress of 10.0 kg/mm². The emphasis was on interrupted tests so that the microstructural changes taking place during creep could be observed.

Helium reduced the rupture life and ductility of the alloy. It also increased the secondary creep rate and changed the failure mode from transgranular to intergranular, but did not affect the intragranular deformation mechanisms.

Helium is swept into the grain boundaries by dislocation motion very early in the creep process. This increases grain-boundary sliding, as shown by separate tensile test results. The helium in the grain boundaries agglomerates into bubbles, the largest being at carbide-matrix interfaces. The effect of the sliding and the bubbles is to cause early nucleation, and subsequent rapid growth, of intergranular cracks leading to premature failure.

I. INTRODUCTION

Creep properties of the cladding become very important owing to the long anticipated fuel life of fast-breeder reactors. Premature rupturing of the cladding can lead to coolant contamination and early removal of the fuel. This in turn would decrease considerably the economic advantages that the breeder reactor has over other methods of producing power. Thus cladding integrity is essential to the performance of the reactor.

One of the major problems in the high temperature performance of the cladding is that of helium embrittlement. Helium is generated in the lattice of the cladding alloy by the (n, α) reactions of high energy neutrons. When the helium is transported to the grain boundaries, it causes a severe loss in ductility.

We have been studying the effect of helium on several cladding alloy candidates⁽¹⁻⁵⁾ to determine extent and mechanisms of helium embrittlement. Our method has been to irradiate the alloy with α -particles from a cyclotron, perform mechanical tests at elevated temperatures, and then interpret changes in mechanical properties in terms of the microstructure. As a part of this study we have conducted a series of creep tests on Type 316 stainless steel at 700°C with an initial stress of 10.0 kg/mm². The main emphasis was on interrupted tests so that the microstructural changes taking place during creep could be observed.

II. EXPERIMENTAL

A. SAMPLE PREPARATION

Samples (0.23 mm by 1.02 mm with a gage length of 12.7 mm) of Type 316 stainless steel were fabricated from sheet stock and irradiated with α -particles from a cyclotron. The details of the irradiation technique have been reported earlier.⁽¹⁾ The composition of the alloy is given in Table 1. The helium concentration in the samples, as determined by mass spectrometry using an isotopic dilution method,⁽⁶⁾ was 4.0×10^{-5} atom fraction.

TABLE 1
COMPOSITION OF TYPE 316 STAINLESS STEEL
(wt %)

Fe	Cr	Ni	Mo	N	C
bal	17.3	13.6	2.3	0.05	0.06

The heat treatment given the alloy prior to helium injection was 1 hr at 982°C + 8 hr at 760°C. The resulting microstructure consisted of austenitic grains, about 40 μ in size, with $M_{23}C_6$ carbides on the grain boundaries.

B. TESTING

All creep tests were done in vacuum ($\sim 5 \times 10^{-6}$ torr). The samples were heated to 700°C and dead loaded after a 1-hr stabilization period. The sample temperature was maintained to within $\pm 3^\circ\text{C}$ during the tests. All tests were constant load tests with an initial stress of 10.0 kg/mm².

The strain was measured by using two linear-variable-differential transformers (LVDT) attached to the base of the sample support system. The LVDT probes rested on the plate containing the weights for loading the sample so that the strain was measured by movement of the load train. The voltages from the LVDT's were recorded on a data logging system and the readings converted to strain by a computer that also produced a CRT plot of the creep curve.

The conversion constant, from volts to length, for each LVDT was obtained by recording the voltage readings as a function of probe displacement, using a micrometer to measure the changes in distance. These measurements were made with the LVDT's mounted in their final position.

With this particular arrangement, the amount of strain that takes place on loading the samples cannot be measured accurately. This is because (a) the preloading voltage of the LVDT is not the same as the true "zero" point voltage at the instant of loading, and (b) the voltage changes immediately after loading are recorded as averages over the integrating time span of the digital voltmeter. As a result, the true "zero" point cannot be determined by extrapolating the creep curve back to time equals zero.

This uncertainty in the "zero" point produces an error in the absolute value of the strain of about 1% but does not alter the essential shape of the creep curve. As the strain for different samples is to be compared, a standard loading procedure was used so that the relative error between samples would be minimal.

For the samples tested to times less than failure, the heater was removed from around the vacuum retort, the sample allowed to cool to about 150°C, and then the load removed.

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Figure 1. Creep Curves for Samples With and Without Helium Tested to Failure

WITHOUT HELIUM

WITH HELIUM

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Figure 2. Light Photomicrographs of Samples With and Without Helium Tested to Failure. The failure mode is transgranular in the sample without helium and intergranular in the sample containing helium.

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III. RESULTS

A. CREEP TESTS

Figure 1 shows the creep curves for samples tested to failure, with and without helium, at 700°C, at an initial stress of 10.0 kg/mm². The effect of helium is to (1) reduce the rupture life, (2) reduce the strain to failure, and (3) increase the secondary creep rate. This is more apparent by comparing the tabulated data given in Table 2 which also includes interrupted test results. From these data we see that the major reduction in elongation occurs in third stage creep.

B. METALLOGRAPHY

Figure 2 shows light photomicrographs of samples, with and without helium, tested to failure. Note the copious amount of intergranular cracks in the sample containing helium. Cracking is limited to the fracture region (to about 2 mm from the fracture edge) for the control, and extends about three times further down the gage length in the case of the sample with helium. The failure mode has changed from transgranular in the control to intergranular in the specimen with helium. There is also a lack of distortion of the grains in the helium-containing sample in the failure zone and the sample has not necked down in this region as has the control sample.

The dislocation substructure of the two samples is shown in Figures 3 and 4. The control sample, shown in Figure 3, has well developed subgrains, whereas the helium-containing sample exhibits long, well-developed, dislocation walls that do not form subgrains (see Figure 4).

Since the substructure is usually a function of the amount of strain, samples tested to 190 hr with and without helium were examined. Here the strains are about equal and the substructure, shown in Figures 5 and 6, is also about the same, with long dislocation walls starting to form, but no definite cell structure.

New precipitation was observed in both samples with and without helium after testing to about the same amount of strain. The precipitates were $M_{23}C_6$ nucleated in the matrix and sigma phase, generally nucleated on existing $M_{23}C_6$ particles. Figure 7 shows a sigma particle growing on a $M_{23}C_6$ carbide.

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Figure 3. Subgrains in a Sample Without Helium Tested to Failure, Transmission Electron Micrograph

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Figure 4. Dislocation Walls in a Helium-Containing Sample Tested to Failure, Transmission Electron Micrograph

TABLE 2
SUMMARY OF CREEP TESTS AT 700°C IN VACUUM
AT AN INITIAL STRESS OF 10.0 kg/mm²

Sample	Helium (appm)	Time (hr)	Creep Strain (%)				Secondary Strain Rate (10 ⁻⁴ /hr)
			Primary	Secondary	Tertiary	Total	
C-5	0	645	3	11	38	52	4.4
C-6	0	561	4	11	17	32	4.2
C-19	0	120(i)*	3	4	none	7	4.5
C-20	0	120(i)	4	3	none	7	4.9
C-21	0	190(i)	3	7	none	10	4.8
C-22	0	25(i)	2	none	none	2	none
C-23	0	303(i)	4	12	none	16	4.7
C-24	0	70(i)	-	-	none	4	-
C-25	0	405(i)	4	9	7	20	4.3
C-26	0	310(i)	3	8	3	14	4.1
19-8	40	148	2	7	2.0	11	6.3
19-9	40	218	2	7	5	14	5.5
19-16	40	70(i)	-	-	-	5	-
19-17	40	70(i)	-	-	-	5	-
19-20	40	120(i)	1	6	none	7	5.8
19-21	40	120(i)	0	7	none	7	5.3
19-24	40	25(i)	1	1	none	2	5.6
19-25	40	190(i)	1	6	4	11	5.0
19-56	40	5(i)	<1	none	none	<1	-

*(i) means creep test was interrupted at indicated time.

8303

Figure 5. Dislocation Walls in a Helium-Containing Sample Tested for 190 hr

8312

Figure 6. Dislocation Walls in a Sample Without Helium Tested for 190 hr

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Figure 7. Sigma Particle Growing on a M₂₃C₆ Carbide Particle in a Helium-Containing Sample Tested to Failure

The only significant difference in microstructural features between the control and helium-containing samples was the presence of helium bubbles in the matrix and grain boundaries of the latter specimens. Helium bubbles, seen in samples tested for only 5 hr, were about 20 Å in size and located in both matrix and grain boundaries.

In Figure 8, bubbles in a sample tested for 70 hr are seen in the grain boundary, generally at the carbide-matrix interface, the largest being about 300 Å. It should also be noted that the dislocations generally do not appear bowed, indicating that the helium bubbles are not pinning them.

The bubble distribution and maximum size observed did not change significantly between 70 and 120 hr of testing. However, after 190 hr of testing the bubbles were considerably larger. After failure, helium bubbles (or voids) 1000 Å in size and larger were not uncommon.

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Figure 8. Transmission Electron Micrograph of a Helium-Containing Sample, Tested for 70 hr, Showing Helium Bubbles in the Grain Boundary

As an extension of bubble formation, void formation at grain boundaries is accelerated in the helium-containing samples. This is clearly shown in replicas of the various specimens. The sequence in Figure 9 shows the progression of void formation from single voids to multiple, but separate, voids to voids that are so closely spaced as to virtually constitute a complete crack. Note that the voids in Figure 9a are not located at the triple points. This is a general feature of the initial void formation in the helium-containing samples.

Although void formation is well advanced in the helium-containing samples tested for 190 hr, as shown in Figure 9c, the amount of actual grain-boundary separation observed is negligible. However, after failure, the amount of grain-boundary separation seen is considerable, as shown in Figure 2. At 190 hr of testing the amount of strain remaining to failure is, on the average, about 3%. Thus, the large amount of grain boundary separation observed after failure occurs in this final portion of tertiary creep.

Although voids were seen in the specimens with helium after testing for 5 hr, they were extremely rare. To check that these were not artifacts, a sample that was helium injected but not tested was examined and no voids were seen. Voids located at triple points were observed in the control sample after 25 hr of testing (the shortest test for a control sample), also being extremely rare. Thus voids are seen in the primary stage of creep in both control and helium-containing samples.

Void nucleation is much less in the control samples since very few voids were observed even after 405 hr of testing, whereas in the samples containing helium, voids continue to nucleate and grow. They are common after 70 hr of testing, and by 190 hr of testing are so numerous as to link up across many grain boundaries.

In an independent study, it was found that the amount of grain boundary sliding that occurs in tensile testing in a sample containing helium is greater than in a sample without helium. The results are given in Table 3. Both samples were tested to their limit of uniform elongation. The details of the method were reported earlier.⁽⁴⁾ The parameter γ is $(\epsilon_{gb}/\epsilon_{total}) (100)$ which is a measure of the relative amount of strain due to grain-boundary sliding. These results indicate that γ is approximately four times greater in a sample containing helium.

TABLE 3
GRAIN-BOUNDARY SLIDING DATA FOR TYPE 316
STAINLESS STEEL TENSILE TESTED AT 800°C

Helium Concentration (ppma)	Elongation (%)	Elongation Due to Grain- Boundary Sliding (%)	γ (%)
0	9.1	0.89 ± 0.23	9.8 ± 2.5
40	3.6	1.3 ± 0.5	36.0 ± 13

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a. 70 hr

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b. 120 hr

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c. 190 hr

71-A13-27-6

Figure 9. Replica Electron Micrographs of a Helium-Containing Sample, Tested to 70, 120, and 190 hr, Showing the Development of Grain-Boundary Voids

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IV. DISCUSSION

The observed reductions in rupture life and elongation at failure are similar to the results of a number of other investigations⁽⁷⁻¹⁴⁾ on neutron-irradiated alloys. The effect of irradiation on the minimum or secondary creep rate is not clear at this time. It has been reported that the minimum creep rate has increased,^(9,10) decreased,^(10,13) or remained the same^(7,8,11,12) when compared to unirradiated controls. Recently, Ring and Rieger⁽¹⁵⁾ have reported that the minimum creep rate is a function of the stress at a particular test temperature. Their results showed minimum creep rates that were both greater than, and less than the control, depending on stress.

The increase in the secondary creep rate in the present case may be explained on the basis of grain-boundary sliding. The results of a separate study, given in Table 3, show that the relative amount of grain-boundary sliding is increased. From these results we infer that helium will enhance grain-boundary sliding during creep at 700°C.

The similarity in microstructural details observed in samples with and without helium tested to the same time indicates that the amount of intragranular strain per unit time is independent of helium during primary and secondary creep. Furthermore, Bloom⁽¹⁴⁾ concluded that neutron irradiation did not affect the intragranular deformation modes of Type 304 stainless steel, creep tested at 650°C, whereas it did affect the early initiation and subsequent growth of intergranular cracks. Therefore, the observed increase in the secondary creep rate in samples containing helium is probably due to an increase in grain-boundary sliding.

Garafalo⁽¹⁶⁾ has pointed out that the evidence is strong that grain-boundary sliding controls intergranular crack initiation; and that crack growth may be due to a number of mechanisms, including grain-boundary sliding. Thus increased grain-boundary sliding in the helium-containing samples is probably responsible for the early formation and rapid growth of voids observed in the present study.

Our results indicate that dislocations sweep helium into the grain boundaries very early in the primary stage, within the first 5 hr of creep. This is in agreement with a previous study where it was shown that a small amount of strain at 760°C produced a large amount of helium bubbles in the matrix and grain boundaries of the alloy, whereas, aging for 4 hr at this temperature did not produce any visible bubbles.⁽¹⁾ The creep tests reported here were done at an even lower temperature, 700°C, thereby slowing further the diffusion process. Thus we believe that dislocation motion is responsible for transporting the helium to the grain boundaries.

The largest bubbles are generally located at carbide-matrix interfaces and, under the combined action of the applied stress and the accelerated grain-boundary sliding, rapidly grow into observable grain-boundary voids. The voids continue to nucleate, grow with increasing strain, finally linking up along the grain boundary. This severely reduces the load-carrying cross sectional area of the sample thereby increasing the stress on the remainder of the sample. Finally, grain-boundary separation starts to take place and intergranular failure occurs by crack propagation.

V. CONCLUSIONS

Helium reduces the rupture life and ductility of Type 316 stainless steel, creep tested at 700°C, under constant load conditions, with an initial stress of 10.0 kg/mm². It also increases the secondary creep rate and changes the failure mode from transgranular to intergranular.

The intragranular deformation processes are not affected by helium in the matrix, as evidenced by dislocation substructure.

Helium is swept into the grain boundaries by dislocation motion very early in the creep process. This leads to an increase in grain-boundary sliding, as shown by tensile tested samples, and can account for the increase in secondary creep rate. The helium in grain boundaries agglomerates into bubbles.

The increase in grain-boundary sliding combined with helium bubbles in the grain boundaries causes the early nucleation and rapid growth of voids at grain-boundary carbides. The voids grow becoming intergranular cracks with additional strain. Grain boundary cracking proceeds rapidly during tertiary creep causing premature failure.

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