MASTER**FATIGUE BEHAVIOR OF IRRADIATED HELIUM-CONTAINING FERRITIC
STEELS FOR FUSION REACTOR APPLICATIONS***

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P.O. Box X, Oak Ridge, TN 37831 U.S.A.**Abstract**

The martensitic alloys 12Cr-1MoVW and 9Cr-1MoVNB have been irradiated in the High Flux Isotope Reactor (HFIR) and subsequently tested in fatigue. In order to achieve helium levels characteristic of fusion reactors, the 12Cr-1MoVW was doped with 1 and 2% Ni, resulting in helium levels of 210 and 410 at. ppm at damage levels of 25 dpa. The 9Cr-1MoVNB was irradiated to a damage level of 3 dpa and contained <5 at. ppm He. Irradiations were carried out at 55°C and testing at 22°C. No significant changes were found in 9Cr-1MoVNB upon irradiation at this damage level, but effects that could possibly be attributed to helium were found in 12Cr-1MoVW. Levels of 210 and 410 at. ppm He produced cyclic strengthening of 29 and 34% over unirradiated nickel-doped materials, respectively. This cyclic hardening attributable largely to helium resulted in degradation of the cyclic life. However, the fatigue life remained comparable to or better than unirradiated 20%-cold-worked 316 stainless steel.

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Introduction

Ferritic, or martensitic, steels have attracted attention for fusion reactor applications due to their radiation resistant properties. Variations of alloys based on 12Cr-1Mo and 9Cr-1Mo compositions have been shown to exhibit low swelling and low irradiation creep rates.¹ These alloys have further appeal for fusion reactor first wall and blanket structures because they have higher thermal conductivity and lower thermal expansion than austenitic stainless steels. The consequence is reduced thermal stress and increased thermal fatigue life, provided the unirradiated fatigue properties remain unchanged by irradiation. Transmutation-induced helium could alter fatigue life, and it is this point that is addressed in this paper.

The helium production rate in a fusion reactor is dependent on reactor design as well as alloy composition. A range of expected helium levels for a stainless steel wall and a lithium-cooled blanket is given in Fig. 1 based on calculations by Gabriel et al.² Figure 1 also shows the helium concentrations generated in ferritic and austenitic alloys as a function of dpa for several experimental reactors: the Oak Ridge Research Reactor (ORR), High Flux Isotope Reactor (HFIR), and Experimental Breeder Reactor (EBR-II). The helium formed in these reactors results primarily from the reaction $^{58}\text{Ni}(n, \gamma)$ followed by $^{59}\text{Ni}(n, \alpha)$, both of which occur with thermal neutrons. In a given reactor, the helium production rate can be chosen by adjusting the nickel content. This approach was used in the present research, where the nickel concentration of two ferritic steels was varied from 0.5 to 2%.

As can be seen in Fig. 1, an alloy with 2% wt Ni results in a He:dpa ratio after HFIR irradiation comparable with that of a fusion reactor.

Experimental Procedure

In this investigation HFIR irradiation of fatigue specimens was followed by low cycle fatigue testing to evaluate the effect of neutron irradiation and helium on the fatigue life of the alloys.

Two fusion reactor candidate alloys were studied: the Sandvik alloy HT-9, often designated 12Cr-1MoVW, which contains nominally 0.5% Ni, and 9Cr-1MoVNb. Three electroslog-remelted (ESR) heats of 12Cr-1MoVW and one ESR heat of 9Cr-1MoVNb were prepared by Combustion Engineering, Inc. The compositions appear in Table I. One 12Cr-1MoVW alloy (HT-3587) was prepared to match the composition of commercial HT-9. The remaining two were doped with 1 and 2% Ni. A fourth ESR heat (91353) prepared by Carpenter Technology Corp. was also irradiated; this heat has been tested extensively in the United States breeder reactor program. One heat of 9Cr-1MoVNb was studied but not doped with nickel.

The specimens were normalized and tempered using the heat treatments shown in Table II. The result was a tempered martensite structure described by Klueh et al.^{3,4}

The specimens were encapsulated in aluminum tubes that were hydrostatically collapsed onto the specimens. The sealed tubes were irradiated in the HFIR with reactor coolant in direct contact with the collapsed tubes. The 12Cr-1MoVW type alloys were irradiated in an irradiation vehicle designated HFIR-MFE-T1 which was irradiated in the peripheral target position of the HFIR to a fluence of 1.4 to 3.4×10^{26} neutrons/m² ($E > 0.1$ MeV) corresponding to a damage level of

10 to 25 dpa. The 9Cr-1MoVNB was irradiated in HFIR-MFE-RB1 which was irradiated in the beryllium reflector of the HFIR to a fluence of 0.38 to 0.51×10^{26} neutrons/m² ($E > 0.1$ MeV) corresponding to a damage level of 2.5 to 3.3 dpa. The displacement damage and helium levels are provided in Table III.

The fatigue specimens were of the miniature hourglass type with a minimum diameter of 3 mm and 45.2 mm in length. The specimens incorporated threaded ends with conical guides to aid in alignment. Further details are provided by Liu and Grossbeck.⁵ The specimens were tested on a fully remote servo-hydraulic fatigue testing system installed in a radiation hot cell. Strain was measured by a diametral extensometer. The tests were performed at room temperature in air. A triangular strain versus time function beginning with compression at a strain rate of $4 \times 10^{-3} \text{ s}^{-1}$ was used. For low-cycle fatigue (total strain range $> 0.5\%$), tests were controlled on the basis of axial strain calculated by a strain computer from diametral strain, which was measured directly at the minimum gage section. For high-cycle tests, the same strain control was used until a stable hysteresis loop was achieved, at which time control was switched to load, and the frequency was increased by a factor of 10 to reduce test duration. Both the irradiated and unirradiated specimens were loaded remotely using the same procedure in order to avoid differences in alignment.

Results

Results for the 12Cr-1MoVW type alloys appear in Table III and Figs. 2 and 3. Figure 2 is a plot of cyclic stress as a function of

total strain range. Stress range was measured at half the cyclic life where a stable hysteresis loop had been achieved. Each point on the curves represents a separate specimen. For unirradiated 12Cr-1MoVW (0.5% Ni), both heats were plotted on the same curve and not separately identified since the results are indistinguishable. The curve for the irradiated 12Cr-1MoVW falls above the unirradiated curve as a result of the expected radiation hardening at low temperatures. Although only one specimen for each nickel-doped alloy was irradiated, the results were consistent and were, therefore, plotted in Fig. 2 along with corresponding unirradiated specimens. The most important comparison to be made is that between the irradiated specimens and their corresponding unirradiated counterparts. The strength of the undoped 12Cr-1MoVW increased 14% upon irradiation, but the 1% Ni-doped material increased 29% in strength upon irradiation and the 2% Ni-doped material increased 34% in strength.

The curves of total strain range as a function of cyclic life are plotted in Fig. 3. Although data are sparse, where possible the data were fit to an equation of the form (using the method of least squares),

$$\Delta\epsilon_T = AN_f^{-0.12} + BN_f^{-b}, \quad (1)$$

where

$\Delta\epsilon_T$ = total strain range,

N_f = cycles to failure,

A, B, b = constants.

The results appear in Table IV.

The curves for both heats of 12Cr-1MoVW, which appear in Fig. 3, show heat 91353 to have a slightly longer fatigue life than heat 3587 in the unirradiated condition. The irradiated 12Cr-1MoVW shows essentially the same cyclic life as the unirradiated material. The nickel-doped alloys, although based on one specimen each, show fatigue lives similar to undoped material in the unirradiated condition. The irradiated nickel-doped alloys exhibited shorter fatigue lives than their unirradiated counterparts, but the differences are small.

It should be noted that, despite the observed effects, the fatigue life of all of the variants of 12Cr-1MoVW remained as good or better than that of unirradiated 20%-cold-worked type 316 stainless steel (20% C.W. 316). Cyclic life of 20% C.W. 316 at room temperature is also plotted in Fig. 3 for comparison purposes.

The cyclic stress curves for 9Cr-1MoVNb appear in Fig. 4. Again, radiation strengthening is evident, but the effect is slight because of the low damage level of only 2 to 4 dpa. Nonetheless, this result is significant since these damage levels are two or three orders of magnitude greater than the embrittling damage level for pressure vessel steels. The cyclic life curves in Fig. 5 also show only a small effect of irradiation. The data for fatigue life of this alloy were again fit to a power law equation of the form of Eq. (1). The results also appear in Table IV. This alloy behaves similarly to unirradiated 20% C.W. 316 but slightly worse than 12Cr-1MoVW.

Discussion

The blanket and perhaps the first wall of any fusion reactor that produces useful energy must operate at elevated temperatures. However, in the present series of scoping experiments, the material was irradiated at only 55°C and tested at room temperature. These temperatures were chosen because experimental research on pressure vessel steels, consisting of mostly fracture toughness-type-tests, indicated that low-temperature irradiation and testing would provide the most adverse conditions.⁶ The lowest temperature attainable in the HFIR is 55°C. Fatigue testing was done at 22°C since no significant differences in performance were expected between room temperature and 55°C.

Normal scatter in fatigue life data makes it difficult to draw conclusions from the sparse data presented. However, cyclic stress-strain curves are more reproducible and normally yield consistent results. Therefore, they will be discussed first.

The cyclic strength for the 12Cr-1MoVW type steels (Fig. 2) does not differ significantly between the two heats of undoped 12Cr-1MoVW. The unirradiated nickel-doped material shows a small increase in cyclic strength for the 1% Ni-doped alloy and a 26% increase for the 2% Ni-doped alloy. The effects of nickel on the unirradiated steels have been discussed by Klueh et al.^{3,4} They found two primary effects: lowering of the A_{c1} temperature, and lowering the martensite start (M_s) and finish (M_f) temperature. If the nickel-doped steel is tempered above the A_{c1} temperature, partial austenitization occurs, which leads to martensite formation

upon cooling. This leads to untempered martensite in the nickel-doped material. If the new M_f temperature is below room temperature, the alloy will contain retained austenite following normalization. When a steel with retained austenite is tempered, precipitation of carbon results in an increase in M_s and M_f . Upon cooling, the austenite can transform to martensite.

To prevent austenite formation in tempering, the 2% Ni-doped material was tempered at 700°C determined by Klueh et al.⁴ to be below the A_{c1} temperature. For the 1% Ni-doped alloy, the previously developed tempering temperature of 780°C was determined to be below the A_{c1} temperature and was therefore used. The M_s and M_f temperatures were determined by Klueh et al.³ to remain above room temperature even in the 2% Ni-doped alloy so that no difficulties were expected. The problem remaining is to develop a heat treatment that produces equivalent microstructures for the doped and undoped alloys. The 5 h heat treatment at 700°C was developed for this purpose, but the high cyclic strength is evidence that this was not fully achieved. Apparently the tempering resulting from the 700°C treatment was not as complete as that achieved by the 780°C heat treatment.^{3, 4}

Keeping in mind that the unirradiated microstructures are similar but not the same, the effects of irradiation will now be addressed. In undoped 12Cr-1MoVW, irradiation strengthening is apparent. The effect of irradiation-induced defects on hardening at low temperatures is well known, and the increase in cyclic strength is expected. The increase in cyclic strength at 1% total strain is 14% upon irradiation. To evaluate the effect of irradiation and helium, the ratio of the cyclic strength for

irradiated material to that for unirradiated material will be examined. For 1% Ni-doped material the ratio is 1.29 and for 2% Ni-doped material it is 1.34. These are to be compared with 1.14 for the undoped alloy. If we can assume that the effects of irradiation for the three alloys are similar and that strengthening by displacement damage is the same, then the additional strengthening observed in the nickel-doped alloys is attributable to helium, 210 and 410 at. ppm for 1% and 2% Ni, respectively. Clearly there are insufficient data to draw strong conclusions, but the additional strengthening observed in the nickel-doped alloys is significant, and the starting microstructures of the doped and undoped alloys appear similar.^{3,4} The differences in microstructure, evidenced by the differences in cyclic strength, are expected to become even less significant upon irradiation. If the increments in cyclic strength are due to hardening by helium, then the increment in strength should increase as $C^{1/2}$ where C is the concentration of helium.⁷ Since there is much hardening already present from dislocation loops and point defects as well as dislocation networks from fatigue, a quantitative approach has limited physical meaning. However, if an equation of the form,

$$\Delta\sigma = A(N + C)^{1/2} ,$$

where

$\Delta\sigma$ = increment in cyclic stress in MPa,

C = helium concentration in at. ppm,

N, A = constants,

is used then the data may be used to determine N and A. The result is

$$\Delta\sigma = 23(18 + C)^{1/2} .$$

This empirical equation fits the data remarkably well, lending support that the hardening is caused by point defects such as an interstitial solute, in particular helium. The constant N represents an artificial defect concentration producing strengthening equivalent to the strengthening resulting from all other sources besides helium. By using the same value of N for all alloys, it is assumed that the point defect contribution to strengthening is not strongly influenced by the differences in the initial microstructures.

The cyclic strength observations may now be used to interpret the cyclic life curves of Fig. 3. The curves for the two heats of 12Cr-1MoVW show a heat-to-heat variation that cannot be attributed to strength since cyclic strengths were similar. This variation should be used as a guide and serve as a caution in evaluating the significance of small changes. The decreases in fatigue life observed for the nickel-doped alloys are indeed small and comparable with the heat-to-heat variation. Nonetheless the decrease is consistent with the observed cyclic hardening, the alloy containing more helium having the shorter life. Strengthening is predicted to reduce low-cycle fatigue life.⁸ It is, therefore, likely that the loss of fatigue life is a consequence of hardening from helium. It is also noteworthy that the strengthening resulting from incomplete tempering in the nickel-doped alloy did not adversely affect fatigue life.

The 9Cr-1MoVNb also shows cyclic hardening due to irradiation. However, the low fluence produces a small change, insufficient to produce a measurable effect on cyclic life.

Conclusions

1. The low-cycle fatigue behavior of 12Cr-1MoVW steel is similar to or better than that of 20% C.W. 316 at room temperature. Irradiation to

10 to 25 dpa does not degrade fatigue life below that of unirradiated 20% C.W. AISI 316 stainless steel.

2. Helium concentrations up to 410 at. ppm produce strengthening in 12Cr-1MoVW steel at room temperature; a possible reduction in fatigue life was also observed.

3. The fatigue life of 9Cr-1MoVNb steel is not significantly altered by irradiation to 2 to 4 dpa, although the cyclic life is slightly lower than that of 12Cr-1MoVW steel.

The results indicate that nickel doping may be used to produce fusion reactor levels of helium in martensitic steels. The effects of the nickel are reasonably well understood so that the effects of helium may be separated. The results indicate that the martensitic steels may be used at low temperatures to damage levels of 25 dpa without appreciable degradation of the fatigue life. Nonetheless, since a possible helium effect was observed, it is prudent to perform a similar investigation at elevated temperatures.

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Table I. Compositions of martensitic steels investigated

Element	Alloy Content, wt %				
	12Cr-1MoVW (91353)	12Cr-1MoVW (3587)	12Cr-1MoVW + 1 Ni (3588)	12Cr-1MoVW + 2 Ni (3589)	9Cr-1MoVNb (3590)
C	0.21	0.21	0.20	0.20	0.09
Mn	0.50	0.50	0.47	0.49	0.36
P	0.007	0.011	0.010	0.011	0.008
S	0.003	0.004	0.004	0.004	0.004
Si	0.22	0.18	0.13	0.14	0.08
Ni	0.58	0.43	1.14	2.27	0.11
Cr	11.80	11.99	11.97	11.71	8.62
Mo	1.02	0.93	1.04	1.02	0.98
V	0.32	0.27	0.31	0.31	0.209
Nb		0.018	0.015	0.015	0.063
Ti	0.003	0.003	0.003	0.003	0.002
Co	0.02	0.017	0.015	0.021	0.013
Cu	0.04	0.05	0.05	0.05	0.03
Al	0.028	0.030	0.017	0.028	0.013
B	<0.001	<0.001	<0.001	<0.001	<0.001
W	0.50	0.54	0.53	0.54	0.01
As	<0.001	<0.001	0.002	<0.002	<0.001
Sn	0.01	0.002	0.001	0.002	0.003
Zr		<0.001	<0.001	<0.001	<0.001
N		0.020	0.016	0.017	0.050
O	<0.006	0.005	0.007	0.007	0.007

Table II. Heat treatment of alloys

Alloy	Heat	Heat treatment
12Cr-1MoVW	91353	1038°C — 10 min AC 760°C — 30 min AC
12Cr-1MoVW	3587	1050°C — 0.5 h 780°C — 2.5 h AC
12Cr-1MoVW + 1 Ni	3588	1050°C — 0.5 h 780°C — 2.5 h AC
12Cr-1MoVW + 2Ni	3589	1050°C — 0.5 h 700°C — 5 h AC
9Cr-1MoVNb	3590	1040°C — 0.5 h 760°C — 1 h AC

Table III. Irradiation parameters and fatigue data

Specimen	Alloy	Heat	Damage Level (dpa)	Helium (at. ppm)	Strain Range (%)	Stress Range $\Delta\sigma$ at $N_f/2$ (MPa)	Cycles to Failure (N_f)
SB01	12Cr-1MoVW	3587	0		2.0	1338	1,807
SB11	12Cr-1MoVW	3587	0		1.0	1104	22,814
SB12	12Cr-1MoVW	3587	0		0.5	924	644,665
SB03	12Cr-1MoVW	3587	10	34	1.0	1268	30,908
SB10	12Cr-1MoVW	3587	15	53	0.5	993	227,652
SJ02	12Cr-1MoVW	91353	0		2.0	1324	4,029
SJ01	12Cr-1MoVW	91353	0		1.0	1117	45,762
SJ07	12Cr-1MoVW	91353	0		0.5	972	719,759
SC02	12Cr-1MoVW-1Ni	3588	0		1.0	1172	26,362
SC04	12Cr-1MoVW-1Ni	3588	24	210	1.0	1517	15,581
SD02	12Cr-1MoVW-2Ni	3589	0		1.0	1393	22,010
SD01	12Cr-1MoVW-2Ni	3589	25	410	1.0	1862	10,539
TA17	9Cr-1MoVNb	3590	0		2.0	1186	1,315
TA14	9Cr-1MoVNb	3590	0		0.5	883	204,235
TA19	9Cr-1MoVNb	3590	0		0.3	641	4,983,360
TA20	9Cr-1MoVNb	3590	3.3	4	1.0	1089	13,225
TA09	9Cr-1MoVNb	3590	2.5	2	0.5	938	247,907
TA11	9Cr-1MoVNb	3590	3.3	4	0.3	703	17,859,711

Table IV. Relations for room temperature fatigue life

Alloy	Condition	Equation
12Cr-1MoVW	Unirradiated	$\Delta\epsilon_T = 0.9 N_f^{-0.12} + 15.7 N_f^{-0.29}$
9Cr-1MoVNb	Unirradiated	$\Delta\epsilon_T = 1.82 N_f^{-0.12} + 59.6 N_f^{-0.54}$
9Cr-1MoVNb	Irradiated	$\Delta\epsilon_T = 2.11 N_f^{-0.12} + 91.7 N_f^{-0.6}$
AISI 316	Unirradiated	$\Delta\epsilon_T = 2.25 N_f^{-0.12} + 134 N_f^{-0.68}$

FIGURES

Fig. 1. Helium as a function of displacement damage for alloys with varying levels of nickel irradiated in experimental reactors. The cross-hatched portion shows levels expected in a tokamak fusion reactor.

Fig. 2. Cyclic stress-strain curve for 12Cr-1MoVW type alloys tested at room temperature showing effects of irradiation and helium. Each point represents an individual specimen.

Fig. 3. Cyclic life of 12Cr-1MoVW type alloys tested at room temperature showing effects of irradiation and helium. A curve for 20%-cold-worked type 316 stainless steel is shown for comparison.

Fig. 4. Cyclic stress-strain curve for 9Cr-1MoVNb tested at room temperature showing effects of irradiation. Each datum represents an individual specimen.

Fig. 5. Cyclic life of 9Cr-1MoVNb tested at room temperature showing effects of irradiation. A curve for 20%-cold-worked type 316 stainless steel is shown for comparison.