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The Influence of Helium on Mechanical Properties of Model Austenitic Alloys, Determined Using ^{59}Ni Isotopic Tailoring and Fast Reactor Irradiation

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THE INFLUENCE OF HELIUM ON MECHANICAL PROPERTIES OF MODEL AUSTENITIC ALLOYS, DETERMINED USING ^{59}Ni ISOTOPIC TAILORING AND FAST REACTOR IRRADIATION - M. L. Hamilton and F. A. Garner, Pacific Northwest Laboratory;[a] B. M. Oliver, Rockwell International.

OBJECTIVE

The objective of this effort is to study the separate and synergistic effects of helium and other important variables on the evolution of microstructure and macroscopic properties during irradiation of structural metals.

SUMMARY

Tensile testing and microscopy continue on specimens removed from the first, second and third discharges of the ^{59}Ni isotopic doping experiment. The results to date indicate that helium/dpa ratios typical of fusion reactors (4 to 19 appm/dpa) do not lead to significant changes in the yield strength of model Fe-Cr-Ni alloys. Measurements of helium generated in undoped specimens from the second and third discharges show that the helium/dpa ratio increases during irradiation in FFTF due to the production of ^{59}Ni. In specimens doped with ^{59}Ni prior to irradiation, the helium/dpa ratio can increase, decrease or remain the same during the second irradiation interval. This behavior occurs because the cross sections for the production and burnout of ^{59}Ni are very sensitive to core location and the nature of neighboring components.

PROGRESS AND STATUS

Introduction

Until recently it has been impossible to conduct experiments in which spectrum-related parameters such as helium/dpa ratio were varied without also accepting variations in other important parameters such as displacement rate and temperature history. A technique currently being used, however, allows the study of the influence of helium alone on density change, microstructural evolution and mechanical properties. This technique utilizes isotopic tailoring to vary the helium production rate without introducing changes in neutron spectrum or displacement rate.[1,2] By producing alloys whose only difference is the presence or absence of ^{59}Ni, an isotope that does not occur naturally, and irradiating doped and undoped specimens side by side in the appropriate reactor spectra, it is possible to generate substantial variations in He/dpa ratio without varying any other important parameter.

A particular advantage of isotopic doping experiments is that one need not be concerned with the details of temperature history, which is now known to heavily influence the outcome of some fission-fusion correlation experiments.[3] Since both doped and undoped specimens are irradiated side by side, the primary variable is only the helium/dpa ratio. The production rate of helium in doped specimens is also nearly constant throughout the experiment, providing that no changes occur in the neutron environment.

This report addresses the tensile data obtained from a subset of the specimens irradiated in a larger experiment currently being conducted in the Fast Flux Test Facility (FFTF) utilizing the Materials Open Test Assembly (MOTA). A previous report addressed the results of the first discharge of this experiment from FFTF; this report includes the first results of the second and third discharges.[4] The results of helium measurements from the second and third discharges are also presented in this report.

Experimental Details

The alloys employed in this study were nominally Fe-15Cr-25Ni, Fe-15Cr-25Ni-0.04P and Fe-15Cr-45Ni (wt%) in both the cold worked and annealed conditions. These alloys were chosen to complement those in several earlier studies, one in the Experimental Breeder Reactor-II (EBR-II), designated the AD-1 experiment,[5,6] and another conducted in the Oak Ridge Research (ORR) Reactor, designated MFE-4.[6] The acquisition of the ^{59}Ni, the production of the ^{59}Ni-doped tensile specimens and their irradiation conditions are described in Reference 1. Microscopy disks were also prepared and irradiated; the results of examination of the first discharge of these specimens are described in detail in References 7 and 8. The miniature tensile specimens measured 5.1, 1.0 mm and 0.25 mm in gauge length, width and thickness, respectively. They were tested at room temperature at a strain rate of 4.7×10^{-4} sec^{-1} in a horizontal test frame described in Reference 9. Yield strengths were determined at 0.2% offset. In some cases more than one tensile specimen was tested, but the majority of tests to date have involved only a single test specimen.

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Results of Tensile Tests

Figure 1 presents a schematic representation of the various irradiation sequences of the ^{59}Ni experiment, each defined by their irradiation temperature and their location in FFTF-MOTA. Tensile tests have been completed on unirradiated specimens of all three alloys in both the annealed and 20% cold worked conditions.

Three tensile tests on unirradiated specimens were conducted for each undoped alloy in both the cold worked and the annealed conditions. Since the availability of doped specimens was rather limited, only one was tested for each combination of doped alloy and thermomechanical starting condition. Figure 2 shows that the range of yield strengths in the undoped specimens is not large and that the single doped specimen in each condition did not exhibit any significant difference in behavior. Also shown in Figure 2 are the yield strengths exhibited by the larger specimens of the unirradiated annealed alloys irradiated in the AD-1 and MFE-4 experiment,[6] demonstrating excellent agreement among the three experiments.

Tests on several subsets of irradiated specimens have also been completed. These subsets are the first two discharges at both 365 and 495°C (i.e., after MOTA-1D and MOTA-1E) and a single discharge at 600°C (after MOTA-1D). The 600°C sequence was not continued to higher radiation exposure due to a large overtemperature event in MOTA-1D, reaching 806°C for fifty minutes. The 495°C sequence also suffered an overtemperature, reaching 629°C for fifty minutes, but was continued in the irradiation sequence along with a replacement sequence at 495°C to explore the impact of the temperature excursion on the experiment. The sequence at 365°C did not experience a temperature excursion.

Figures 3-5 show the yield strengths measured at room temperature of specimens irradiated at nominal temperatures of 365, 495 and 600°C. The width of the error bars at zero dpa corresponds to the variation seen in Figure 2 and provides a basis for determining whether variations observed between doped and undoped specimens are significant compared to the scatter associated with the measurement technique.

The most significant features of the data shown in Figures 3-5 are, first, the relative unimportance of isotopic doping at all three temperatures in determining the yield strength, and second, the convergence at

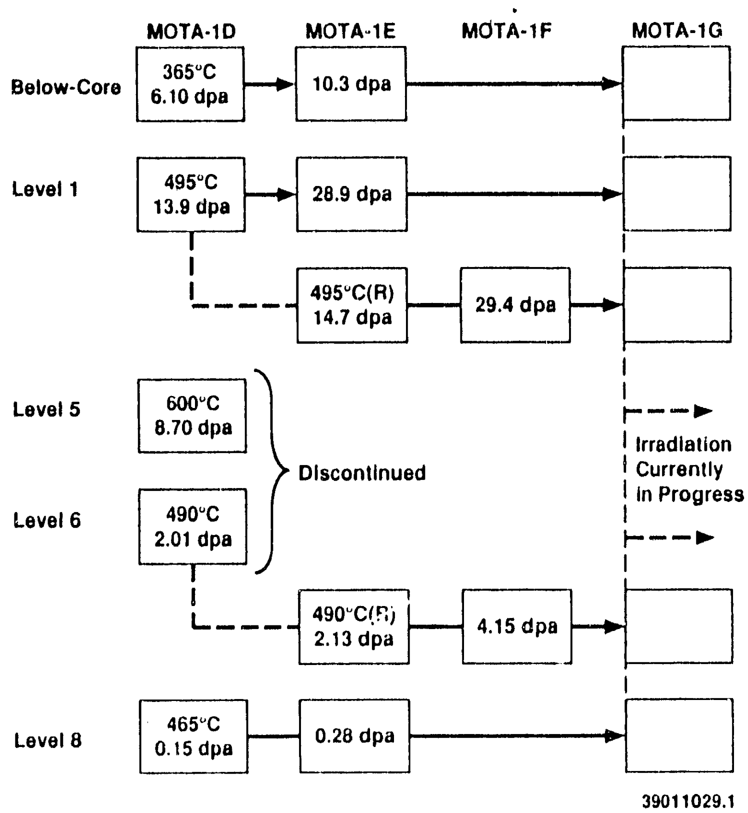

Figure 1. Schematic representation of irradiation sequences for the ^{59}Ni isotopic tailoring experiment. "495°C(R)" and "490°C(R)" refer to the replacement sequences inserted for the original sequences at 495 and 490°C

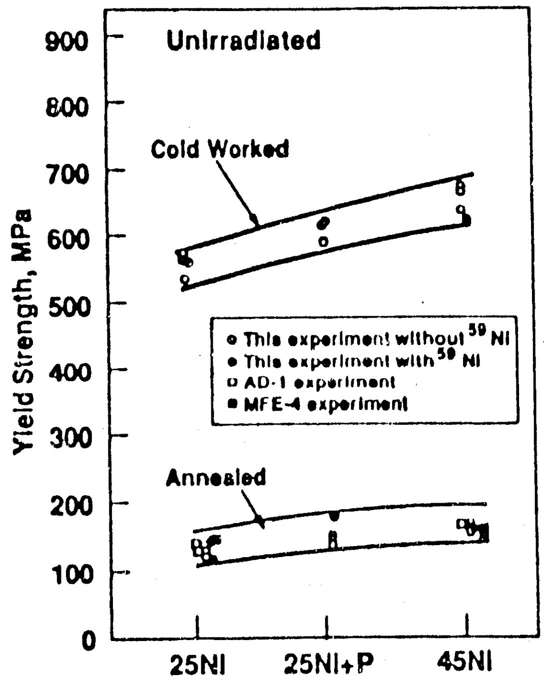

Figure 2. Yield strength data on unirradiated specimens from the FFTF/MOTA ^{59}Ni experiment and two related experiments conducted in other reactors

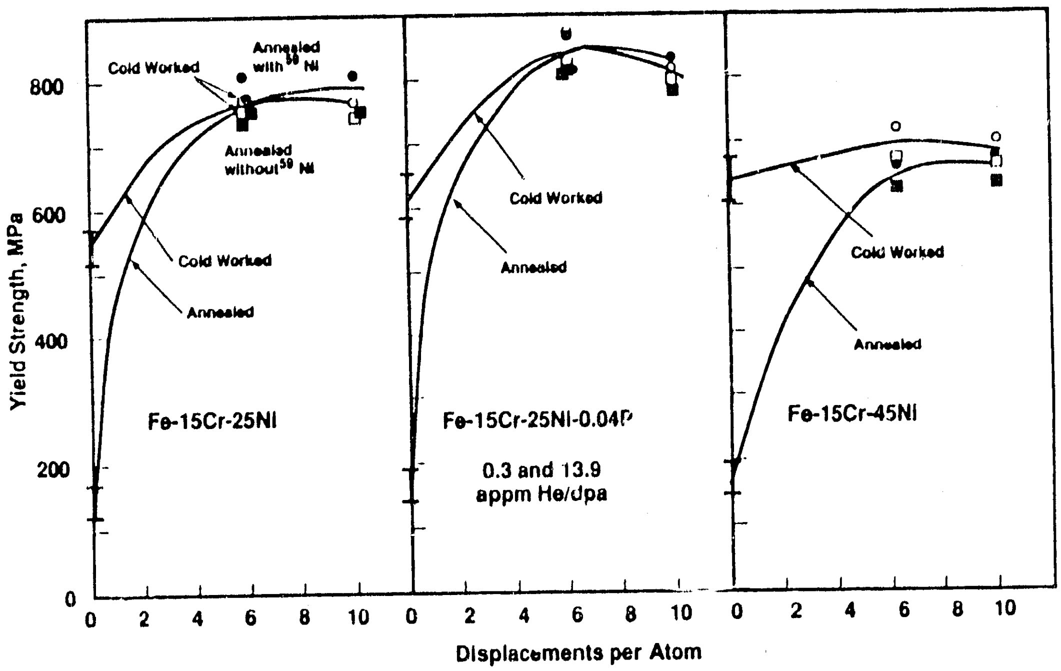

Figure 3. Influence of thermomechanical starting state and isotopic doping on yield strength of three alloys irradiated at 365°C. Open and closed symbols denote cold worked and annealed alloys, respectively; circles denote ^{59}Ni-doped specimens; squares denote undoped alloys

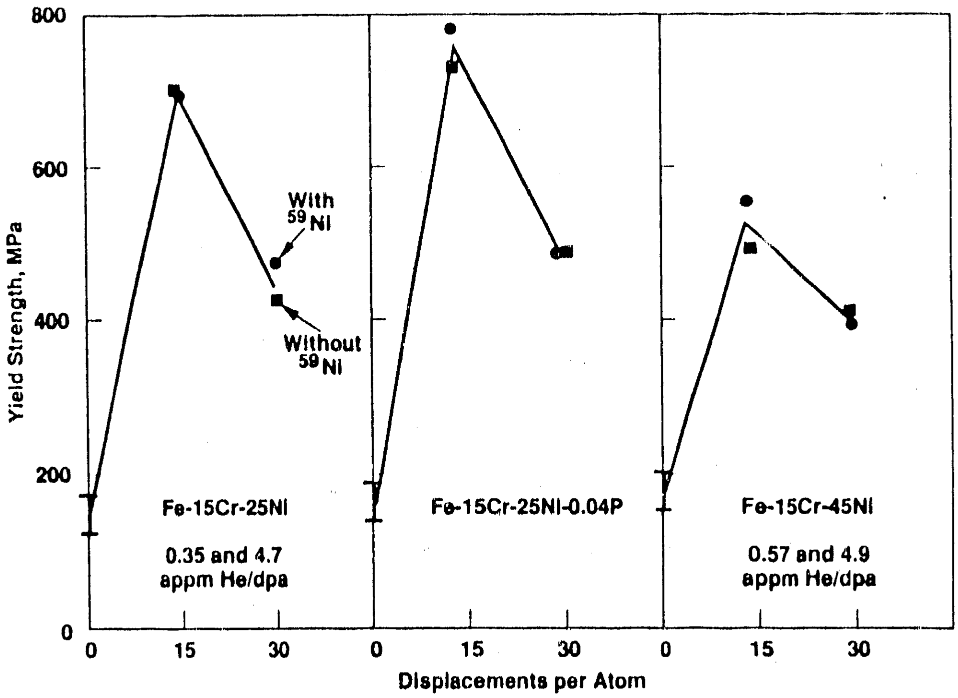

Figure 4. Influence of isotopic doping on yield strength of three annealed model alloys irradiated at 495°C

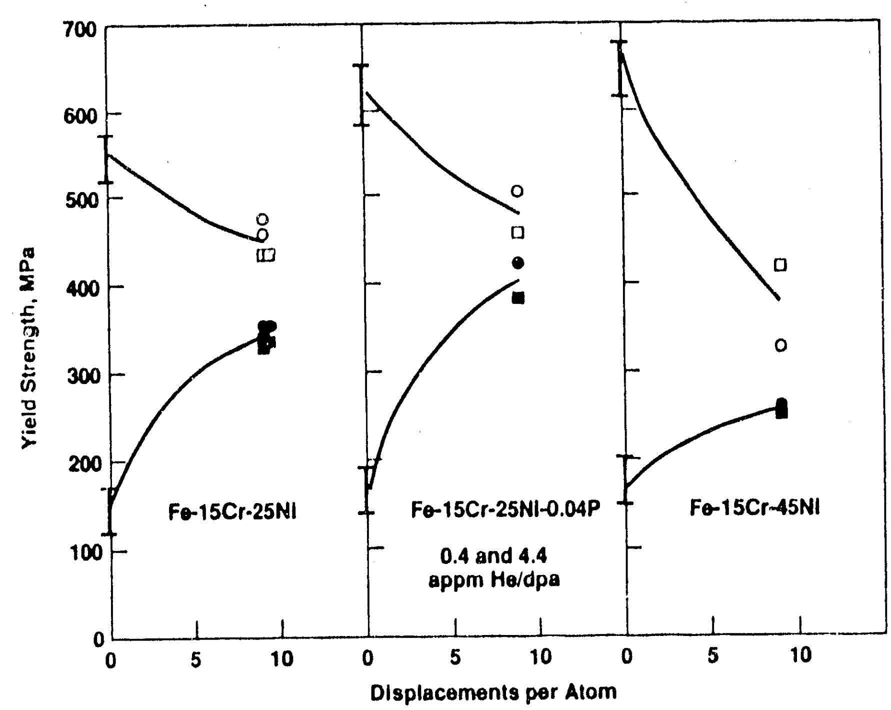

Figure 5. Influence of thermomechanical starting state and isotopic doping on yield strength of three alloys irradiated at 600°C. Open and closed symbols denote cold worked and annealed alloys, respectively; circles denote ^{59}Ni-doped specimens; squares denote undoped alloys.

Table 1
Helium Concentration in Cold Worked TEM Disks from Isotopic Tailoring Experiment

Temperature (°C)	Nickel (wt%)	Doped With ^{59}Ni	dpa	Specimen Mass[a] (mg)	^4He Measured (10^{14} atoms)	Helium Concentration (appm)[b] Measured	Average[c]
365	25	No	10.3	1.937	1.493	7.157	7.12
				2.966	2.263	7.084	±0.05
365	25	Yes	10.3	2.744	48.86	165.3	165
				3.186	56.34	164.2	±1
365	45	No	10.3	2.562	3.375	12.35	12.4
				3.024	4.015	12.44	±0.1
365	45	Yes	10.3	1.996	36.48	171.3	172
				3.339	61.24	171.9	±1
495	25	No	28.9	1.733	2.546	13.64	13.7
				3.410	5.016	13.66	±0.1
495	25	Yes	28.9	2.482	36.22	135.5	136
				3.316	48.46	135.7	±1
495(R)[d]	25	No	14.7	2.644	1.834	6.441	6.50
				3.374	2.385	6.563	±0.09
495(R)	25	Yes	14.7	3.174	23.82	69.68	69.2
				3.554	26.28	68.66	±0.7
495(R)	25	No	29.4	2.087	3.797	16.89	16.8
				2.360	4.247	16.71	±0.1
495(R)	25	Yes	29.4	2.826	36.49	119.9	120
				2.983	38.66	120.3	±1
490(R)	25	No	2.13	2.881	0.1383	0.4457	0.440
				4.212	0.1974	0.4352	±0.007
490(R)	25	Yes	2.13	2.861	6.691	21.71	21.5
				3.643	8.386	21.37	±0.2
490(R)	45	No	2.13	2.709	0.1865	0.6452	0.640
				3.241	0.2197	0.6353	±0.007
490(R)	45	Yes	2.13	2.785	7.296	24.55	24.1
				3.555	8.959	23.62	±0.7
490(R)	25	No	4.15	2.348	0.3590	1.420	1.42
				3.797	0.5808	1.420	±0.00
490(R)	25	Yes	4.15	2.735	16.87	57.27	56.8
				3.177	19.28	56.35	±0.7
465	25	No	0.28	2.825	0.02223	0.07306	0.0727
				3.129	0.02438	0.07235	±0.0005
465	25	Yes	0.28	2.780	5.201	17.37	17.4
				2.746	5.134	17.36	±0.1

(a) Mass uncertainty is ±0.001 mg. Two samples were cut from each original TEM specimen.
(b) Measured helium concentration in atomic parts per million (10^{-6} atom fraction) with respect to the calculated number of atoms in the specimen.
(c) Mean and 1σ standard deviation of duplicate analyses.
(d) R denotes replacement series.

Table 2
Helium Generation Rates Determined in Fe-15Cr-25Ni

Temperature (°C)	Helium Generation Rate (appm/dpa) MOTA-1D Undoped	MOTA-1D Doped	MOTA-1E Undoped	MOTA-1E Doped	MOTA-1F Undoped	MOTA-1F Doped
365	0.30	13.9	0.69[a] (1.26)[b]	16.0[a] (19.1)	no discharge	
495	0.35	4.69	0.47[a] (0.59)	4.71[a] (4.73)	no discharge	
495(R)[c]	not irradiated		0.44	4.71	0.57[a] (0.70)	4.08[a] (3.45)
600	0.40	4.4	not irradiated		not irradiated	
490	0.28	19.8	not irradiated		not irradiated	
490(R)[c]	not irradiated		0.21	10.1	0.34[a] (0.46)	13.7[a] (17.2)
465	0.14	54.3	0.26[a] (0.40)	62.1[a] (71.1)	no discharge	

(a) He/dpa level given is average for two cycle irradiation sequence.
(b) Helium/dpa levels in parentheses represent average values calculated for second MOTA cycle only.
(c) R denotes replacement series.

Table 3
Cumulative Helium Generation Rates Determined in Fe-15Cr-45Ni After Discharge from MOTA-1E

Temperature (°C)	appm/dpa Undoped	appm/dpa Doped	Neutron damage (dpa) MOTA-1D	MOTA-1E	Total
365	1.20	16.7	6.1	4.2	10.3
490R	0.30	11.3	0.01	2.13	2.13

Several consequences of changes in the neutron environment can be seen in the two irradiation sequences at 490°C. First, the helium/dpa ratios for both specimen types in the replacement sequence are somewhat lower than that of the original sequence, probably reflecting differences in the loading of nearby components during the operation of MOTA-1D and MOTA-1E. Second, based on the measured MOTA-1E production rates in undoped Fe-15Cr-25Ni, we would expect the production rate in undoped Fe-15Cr-45Ni to be 0.38 appm/dpa, whereas only 0.30 appm/dpa was measured. The difference between helium levels in the 25Ni and 45Ni doped specimens is also somewhat larger than expected. These observations imply that the 45Ni specimens, which were in a separate packet from the 25Ni specimens, experienced a slightly different spectral environment, even though they were in the same basket. This in turn suggests that radial gradients in spectral parameters must also exist and that the specimens may not have been closely side by side.

The 465°C low fluence sequence was conducted in above core position 8F1 in both MOTA-1D and MOTA-1E. In this case the production rates of both the doped and undoped specimens increased. Note that the production rates in the doped specimens are quite large, 54 and 71 appm/dpa in Fe-15Cr-25Ni in MOTA-1D and MOTA-1E, respectively.

Review of the data in Tables 1 and 2 leads to the following overall conclusions: 1) helium production in undoped specimens containing only natural nickel is greatest for irradiation within the core; 2) significant increases in helium production rate occur with increasing exposure in undoped specimens regardless of location with respect to the core. The percentage increases are greatest for irradiations outside the core. 3) helium production rates in specimens doped with ^{59}Ni dopant prior to irradiation tend to increase also, but are much more sensitive to changes in neutron spectrum arising from changes in positi or changes in neighboring components. In some cases the helium production rate can actually decrease even though undoped specimens in the same reactor position experience increases in helium production. This difference is due to the fact that ^{59}Ni production varies roughly as the second power of the dpa level, while the burn-out of ^{59}Ni is roughly linear with dpa.[14] Both reactions are separately but strongly sensitive to fine details of the neutron spectra.

Status of Other Types of Examination

Electron microscopy has been performed by Prof. J. F. Stubbins (NORCUS assignee at PNL from the University of Illinois) on all twelve specimen conditions of the second discharge at 365°C. Data analysis is now in progress. Density change measurements are in progress on TEM specimens from MOTA-1E at 365°C, 495°C (original) and 495°C (replacement) and the second discharge of the 495°C (replacement) from MOTA-1F.

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FUTURE WORK

This effort will continue, concentrating on microscopy and additional tensile testing.

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