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FUSION MATERIALS SEMIANNUAL PROGRESS REPORT FOR THE PERIOD ENDING

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FOREWORD

This is the sixty-seventh in a series of semiannual technical progress reports on fusion materials science activity supported by the Fusion Energy Sciences Program of the U.S. Department of Energy. It covers the period ending December 31, 2019. This report focuses on research addressing the effects on materials properties and performance of exposure to the neutronic, thermal and chemical environments anticipated in the chambers of fusion experiments and energy systems. This research is a major element of the national effort to establish the materials knowledge base for an economically and environmentally attractive fusion energy source. Research activities on issues related to the interaction of materials with plasmas are reported separately.

The results reported are the products of a national effort involving a number of national laboratories and universities. A large fraction of this work, particularly in relation to fission reactor irradiations, is carried out collaboratively with partners in Japan, Russia, and the European Union. The purpose of this series of reports is to provide a working technical record for the use of program participants, and to provide a means of communicating the efforts of fusion materials scientists to the broader fusion community, both nationally and worldwide.

This report has been compiled by Stephanie Melton, Oak Ridge National Laboratory. Her efforts, and the efforts of the many persons who made technical contributions, are gratefully acknowledged.

Daniel Clark Research Division Office of Fusion Energy Sciences

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1.1 COMPARISON OF CARBIDE- AND CARBONITRIDE-STRENGTHENED CASTABLE NANOSTRUCTURED ALLOYS—L. Tan, T. Graening, X. Hu, W. Zhong, Y. Yang, Y. Katoh (Oak Ridge National Laboratory)

OBJECTIVE

Both carbonitride- and carbide-strengthened castable nanostructured alloys (CNAs) were developed at Oak Ridge National Laboratory (ORNL) as United States (U.S.) reduced-activation ferritic-martensitic (RAFM) steels. The objective is to compare the microstructures and properties of the two types of CNAs to understand their respective advantages or disadvantages over current RAFM steels.

SUMMARY

Two heats of carbonitride- and six heats of carbide-strengthened CNAs were developed and systematically examined. Compared with Eurofer97 and F82H, the CNAs showed greater tensile strength and creep resistance, comparable or greater Charpy impact toughness, but more deuterium retention. Compared with the carbonitride-strengthened CNAs, the carbide-strengthened CNAs showed comparable tensile strength and creep resistance, but a higher density of nanoprecipitates uniformly distributed in the matrix, greater upper-shelf energy (USE) in Charpy impact toughness, less deuterium retention, and less transmutation in the carbides.

PROGRESS AND STATUS

Introduction

Carbonitride is a common type of MX used in the current RAFM steels such as Eurofer97 and F82H. To maximize the MX density in CNAs, carbonitride was boosted by increasing the nitrogen and vanadium concentrations. On the other hand, carbide-strengthened CNAs were developed by eliminating nitrogen and introducing titanium as an alloying element. The two types of CNAs were developed and examined to be compared with the current RAFM steels.

Experimental Procedure

Two heats of carbonitride- and six heats of carbide-strengthened CNAs were designed by computational alloy thermodynamics. The CNAs were fabricated either with vacuum arc melting or vacuum induction melting in laboratory scale heats. The casting heats were homogenized and hot-rolled with the same amount of thickness reduction, followed by normalization at 1150–1170°C for 0.5 h depending on the delta-ferrite formation temperature of the alloys, and then tempering at 750°C for 0.5–1 h with air cooling depending on the final plate thickness. A comprehensive examination was performed for the alloys, which includes microstructure, tensile, creep, Charpy impact, deuterium retention, and radioactivity of alloys and transmutation of phases. This report only shows the examples of microstructure and Charpy impact results.

Results

Figure 1a-b and c-d show the typical microstructures of the carbonitride- and carbide-strengthened CNAs characterized by transmission electron microscopy (TEM) and energy dispersive x-ray spectroscopy (EDS), using alloy CC22 and CNA3 as examples, respectively. Both CC22 and CNA3 have similar lath structures. The EDS maps in Figure 1b from CC22 indicate primarily boundary precipitates, being composed of many V- and N-enriched precipitates with few of them enriched with Ta and some Cr-enriched precipitates. In contrast, Figure 1d of CNA3 shows many ultrafine precipitates in the matrix, together with a coarse Cr-rich M23C6 particle and few fine Ti-rich carbide at the boundary on the left side of this image. Detailed TEM and atom probe tomography analyses indicated that the carbide density in the matrix of CNA3 is by orders of magnitude higher than the rare carbonitrides in the matrix of CC22.



Figure 1. Typical microstructures of (a-b) carbonitride-strengthened CC22 and (c-d) carbide-strengthened CNA3.

The USE as a function of ductile-brittle transition temperature (DBTT) of the eight CNAs are plotted in Figure 2. The literature data with the same transverse-longitudinal (T–L) orientation of Grade 91 (G01) and oxide-dispersion-strengthened (ODS) 14YWT using the same half-size Charpy impact specimens, Eurofer97 (E97), F82H, and CLAM using full-size Charpy impact specimens, and ODS-Eurofer (ODS-EU), PM2000, and MA956 using the KLST type specimens are included for comparison. The absorbed energies were normalized to the nominal fracture volume of the different types of specimens, which would noticeably mitigate the specimen size effect on USE. The CNAs show generally greater USE, except for CNA1, than G91, RAFM steels, and ODS alloys. The carbide-strengthened CNAs generally have greater USE than the carbonitride-strengthened CNA1 and CC22. The DBTT of the CNAs is generally comparable to that of G91 and RAFM steels considering the different specimen sizes.



Figure 2. Charpy impact toughness (USE as a function of DBTT) of the carbonitride (open diamonds) and carbide (filled diamonds) strengthened CNAs compared with G91, F82H, E97, and CLAM.

1.2 POST-IRRADIATION EVALUATION OF FRACTURE TOUGHNESS PROPERTIES FOR EUROFUSION M4CVN BEND BAR SPECIMENS—X. Chen, M. Sokolov, L.N. Clowers, A. Bhattacharya, J.W. Geringer, Y. Katoh (Oak Ridge National Laboratory), T. Graening (Karlsruhe Institute of Technology)

OBJECTIVE

The aim of this task is to measure irradiation embrittlement for EUROFusion M4CVN bend bar specimens machined from ten Eurofer97 steel variants. The neutron irradiation was performed at the High Flux Isotope Reactor (HFIR) of the Oak Ridge National Laboratory (ORNL) with the target irradiation condition of 300°C/2.5 displacements per atom.

SUMMARY

In this project, we performed fracture toughness characterization of ten neutron irradiated Eurofer97 steel variants using precracked miniature multi-notch bend bar specimens based on the Master Curve method in the ASTM E1921 standard. The neutron irradiation was performed in the flux trap position of the HFIR of the ORNL with the nominal irradiation temperature of 300°C and 2.5 displacements per atom. Depending on the irradiation temperature and materials, we observed different degrees of irradiation embrittlement for ten Eurofer97 variants. The upper shift in the Master Curve reference temperature ΔT_{0Q} vs. the increase in Vickers microhardness values showed a liner relationship for only a few materials indicating different irradiation responses of the Eurofer97 variants.

PROGRESS AND STATUS

Introduction

Eurofer97 is the European reference reduced activation ferritic/martensitic (RAFM) steel for the first wall and blanket applications of the DEMO fusion reactor [1-3]. It has favorable properties for fusion applications such as lower radioactivity, superior swelling resistance, and good thermal conductivity [4] and poses enough fracture toughness at the normalized and tempered condition. However, the harsh environment of a fusion reactor, such as neutron irradiation and He/H damage, results in significant degradation of Eurofer97 fracture toughness. Therefore, understanding the Eurofer97 fracture toughness behavior is critical to ensure the long-term safe operation of the fusion reactor. This is especially true for characterizing test reactor irradiated specimens which are usually much smaller than standard size specimens due to limited volume of irradiation facilities and advantages with testing small size specimens, such as lower radioactivity and more accurate control of irradiation temperatures. In this study, we have developed a small specimen testing technique using pre-cracked miniature multi-notch bend bar specimens (referred to as M4CVN hereafter) based on the Master Curve method in the ASTM E1921 standard [5]. Under the framework of EUROfusion, we performed fracture toughness characterization on ten Eurofer97 variants before and after neutron irradiation. The irradiation was performed at the HFIR of the ORNL using rabbit capsules in the flux trap position. The nominal irradiation condition was 300°C and 2.5 displacements per atom (dpa). This paper describes the transition fracture toughness properties of the ten materials with complementary Vickers microhardness results.

Experimental Procedure

Ten Eurofer97 variants (code names: E, H, I, J, K, L, M, N, O, and P) were used for testing. The materials compositions and heat treatment conditions (listed in Table 1 and Table 2, respectively) were intentionally varied to study the effects of these two factors on mechanical and microstructural properties. It is worth noting that materials E and M are the standard Eurofer97 heat and the heat treatment condition for material E also represents the standard heat treatment for Eurofer97.

	Cr	С	Mn	V	Ν	W	Та	Si
Е	8.8	.11	.53	.20	.019	1.1	.12	.04
Н	8.7	.06	.02	.35	.047	1.1	.10	.04
I	8.7	.11	.02	.35	.042	1.1	.09	.04
J	9.0	.11	.39	.22	.022	1.1	.11	<.04
Κ	7.8	.02	<.03	.22	.022	1.0	.13	<.04
L	9.1	.11	.54	.20	.038	1.1	.12	.03
Μ	8.8	.11	.53	.20	.019	1.1	.12	.04
Ν	9.0	.09	.11	<.05	.002	1.0	.09	.04
0	8.8	.06	.50	.30	.070	1.0	.05	.15
Ρ	8.7	.11	.02	.20	.045	1.1	.09	.03

Table 1	Chemical	compositions	of ten	Eurofer97	variants	(wt%)
	. Onernicai	compositions	OF LOT	Luidicion	vananto	(** 1 / 0 /

Table 2. Heat treatment conditions for ten Eurofer97 variants

E*	980°C/0.5h + AQ + 760°C + AC
Н	1000°C/0.5h + WQ + 820°C + AC
Ι	1000°C/0.5h + WQ + 820°C + AC
J	1250°C/1h + rolling to a final rolling temperature of 850°C in 6 rolling steps with a reduction of 20-
	30% for each step + AC + 880°C/0.5h + WQ + 750°C/2h + AC
ĸ	1250°C/1h + rolling to a final rolling temperature of 850°C in 6 rolling steps with a reduction of 20-
IX.	30% for each step + AC + 1050°C/0.25h + WQ + 675°C/1.5h + AC
L	1150°C/0.5h + AQ + 700°C + AC
М	1020°C/0.5h + AQ + 1020°C/0.5h + AQ + 760°C/1.5h + AC
Ν	920°C/1.5h + AQ + 920°C/1.5h + AQ + 760°C/1h + AC
0	1080°C/1h, cooling to 650°C and rolling to 40% reduction + 760°C/1h + AC
Ρ	1000°C/0.5h + WQ + 820°C + AC

AQ: air quench; WQ: water quench; AC: air cooled

*E had experienced unknown heat treatment prior to the shown heat treatment.

Five rabbit capsules were used to irradiate ten Eurofer97 variants. Each capsule contained four M4CVN bend bar specimens machined from two Eurofer97 variants and four SiC passive thermometry specimens which were in direct contact with the bend bar specimens for irradiation temperature monitoring. The method for irradiation temperature measurements using a SiC thermometry specimen has been described in detail by Campbell et al. in Reference [6].

M4CVN specimens with a dimension of 45 (length) x 3.3 (width) x 1.65 (thickness) mm³ were used for Master Curve fracture toughness testing. The specimen was specifically developed within the ORNL Fusion Materials Program and has four notches per specimen. Despite its small size, the M4CVN specimen follows the same size ratio of a bend bar specimen in ASTM E1921. Due to shared loading portions between neighboring notches, the M4CVN specimen consumes significantly less material than a standard single notch bend bar specimen and is favorable for neutron irradiation testing. Each notch on the M4CVN specimen was fatigue precracked to a/W ratio of ~0.5 before testing. Due to the limitation of irradiation cost and volume, 8 notches per material were tested to obtain the Master Curve reference temperature T_{0Q}. The testing procedures, fixture design, and analysis procedures for unirradiated specimens have been described previously in [7, 8]. A few modifications for the test setup have been made to facilitate hot cell testing. As shown in Figure 1, the liquid nitrogen (LN) and air line supplies LN in low temperature testing and hot air for deicing and high temperature testing. The environmental chamber is attached to a linear actuator and can be raised to enclose the specimen for testing and lowered to remove and reload specimens. Since it was not feasible to spot weld thermocouples directly onto the bend bar specimen for the temperature measurement in hot cell testing, indirect temperature measurement was made from thermocouples spot welded to the indenter. To ensure accurate temperature control, temperature calibration was performed between the indenter and a dummy bend bar specimen installed

at the testing position. The dummy specimen had a thermocouple spot welded to it and the temperature readings between the indenter and the dummy specimen were recorded for a temperature range from - 170°C to 75°C to cover the entire testing temperature range.



Figure 1. Hot cell setup for miniature bend bar testing.

Vickers microhardness testing was performed using a Mitutoyo HV-120 hardness tester in the hot cell. We applied 1 kg force with 15 sec dwell time for each measurement. At least four measurements were performed near the end of the initial fatigue precracking location for each notch. The schematic for the Vickers microhardness indentation pattern is illustrated in Figure 2.





Results

Irradiation Temperature

The average irradiation temperatures of ten Eurofer97 variants are shown in Figure 3. Compared with the target irradiation temperature 300°C, most materials achieved reasonable irradiation temperatures considering the complex nature of neutron irradiation. However, materials M and N experienced a much higher irradiation temperature of ~476°C. It is expected that the high irradiation temperature would result in minimum irradiation hardening and embrittlement for materials M and N.



Figure 3. Irradiation temperature measurements for ten Eurofer97 variants. The scatter bands correspond to +/- 20°C.

Vickers Microhardness

The Vickers microhardness results of ten Eurofer97 variants are shown in Figure 4 with the irradiation temperature and unirradiated hardness results overlaid in the same figure. Except for materials M and N which experienced a much higher irradiation temperature, all other materials showed apparent irradiation hardening and the difference in irradiation hardening should mostly be driven by microstructural differences among the materials.



Figure 4. Measured irradiation hardening for ten Eurofer97 variants. The scatter bands correspond to one standard deviation.

Fracture Toughness

The fracture toughness results of ten irradiated Eurofer97 variants are summarized in Tables 3-12. The upper shift in the Master Curve reference temperature ΔT_{oQ} is also listed in the same table.

Notch ID	Test temperature (°C)	K _{Jc} (MPa√m)	1T-K _{Jc} (MPa√m)	Censored (Y/N)	Censored 1T-K _{Jc} value (MPa√m)	T₀₀ (°C)	ΔT₀q (°C)
E030L	-30	44.9	32.6	Ν	NA		
E030LM	-35	45.2	32.7	Ν	NA		
E030RM	-45	42.1	31.1	Ν	NA		
E030R	-20	34.0	27.1	Ν	NA	01	112
E036L	-15	36.4	28.3	Ν	NA	04	113
E036LM	-15	87.9	54.3	Ν	NA		
E036RM	-15	71.7	46.1	N	NA		
E036R	-9	45.5	32.9	N	NA		

Table 3. Fracture toughness of irradiated Eurofer97 material E

Table 4. Fracture toughness of irradiated Eurofer97 material H

Notch ID	Test temperature (°C)	K _{Jc} (MPa√m)	1T-K _{Jc} (MPa√m)	Censored (Y/N)	Censored 1T-K _{Jc} value (MPa√m)	T₀Q (°C)	ΔT _{oQ} (°C)
H000L	-106	346.9	185.0	Υ	58.3		
H000LM	-109	152.0	86.6	Υ	60.5		
H000RM	-58	335.7	179.4	Υ	59.3		
H000R	-95	95.9	58.3	Ν	NA	15	16
H001L	-108	69.6	45.0	Ν	NA	-45	40
H001LM	-122	31.9	26.0	Ν	NA		
H001RM	-120	57.5	38.9	Ν	NA		
H001R	-104	50.7	35.5	Ν	NA		

Table 5. Fracture toughness of irradiated Eurofer97 material I

Notch ID	Test temperature (°C)	K _{Jc} (MPa√m)	1T-K _{Jc} (MPa√m)	Censored (Y/N)	Censored 1T-K _{Jc} value (MPa√m)	T₀Q (°C)	ΔT _{oQ} (°C)
1000L	-112	21.5	20.8	Ν	NA		
1000LM	-118	27.3	23.7	Ν	NA		
1000RM	-105	292.8	157.7	Υ	59.0		
1000R	-100	97.2	59.0	Ν	NA	-32	46
1001L	-99	71.0	45.7	Ν	NA		
1001RM	-99	69.1	44.8	Ν	NA		
1001R	-99	68.1	44.3	Ν	NA		

Notch ID	Test temperature (°C)	K _{Jc} (MPa√m)	1T-K _{Jc} (MPa√m)	Censored (Y/N)	Censored 1T-K _{Jc} value (MPa√m)	T₀Q (°C)	ΔT₀q (°C)
J000L	-125	92.6	56.6	Ν	NA		
J000LM	-109	315.0	168.9	Υ	68.3		
J000RM	-86	348.5	185.8	Υ	66.4		
J000R	-126	60.0	40.2	Ν	NA	50	11
J004L	-125	47.0	33.7	Ν	NA	-56	41
J004LM	-126	36.4	28.3	Ν	NA		
J004RM	-125	88.1	54.4	Ν	NA		
J004R	-125	46.1	33.2	Ν	NA		

Table 6. Fracture toughness of irradiated Eurofer97 material J

 Table 7. Fracture toughness of irradiated Eurofer97 material K

Notch ID	Test temperature (°C)	K _{Jc} (MPa√m)	1T-K _{Jc} (MPa√m)	Censored (Y/N)	Censored 1T-K _{Jc} value (MPa√m)	T₀Q (°C)	ΔT _{oQ} (°C)
K001L	40	47.8	34.0	Ν	NA		
K001LM	25	58.9	39.7	Ν	NA		
K001RM	1	35.5	27.8	Ν	NA		
K001R	60	63.0	41.7	Ν	NA	107	140
K003L	73	79.0	49.8	N	NA	137	149
K003LM	80	160.3	90.8	Υ	70.4		
K003RM	75	68.8	44.6	Ν	NA		
K003R	75	78.4	49.5	N	NA		

Table 8. Fracture toughness of irradiated Eurofer97 material L

Notch ID	Test temperature (°C)	K _{Jc} (MPa√m)	1T-K _{Jc} (MPa√m)	Censored (Y/N)	Censored 1T-K _{Jc} value (MPa√m)	T₀Q (°C)	ΔΤ _{οQ} (°C)
L000L	-34	39.2	29.7	Ν	NA		
LOOOLM	-39	59.4	39.9	Ν	NA		
L000RM	-45	50.0	35.1	Ν	NA		
L000R	-29	41.6	30.9	Ν	NA	10	101
L001L	-25	39.7	30.0	Ν	NA	40	101
L001LM	-19	88.0	54.3	Ν	NA		
L001RM	-19	117.8	69.4	N	NA		
L001R	-19	68.9	44.7	N	NA		

Notch ID	Test temperature (°C)	K _{Jc} (MPa√m)	1T-K _{Jc} (MPa√m)	Censored (Y/N)	Censored 1T-K _{Jc} value (MPa√m)	T₀Q (°C)	ΔT₀Q (°C)
M000L	-136	24.8	22.4	Ν	NA		
M000LM	-120	131.9	76.5	Υ	57.8		
M000RM	-150	19.0	19.5	Ν	NA		
M000R	-126	49.5	34.9	Ν	NA	60	6
M001L	-130	91.7	56.2	Ν	NA	-09	Ø
M001LM	-129	68.6	44.6	Ν	NA		
M001RM	-125	173.6	97.5	Y	60.8		
M001R	-128	99.1	59.9	Y	58.5		

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Table 9. Fracture toughness	of irradiated	Eurofer97	material M

Table 10. Fracture toughness of irradiated Eurofer97 material N

Notch ID	Test temperature (°C)	K _{Jc} (MPa√m)	1T-K _{Jc} (MPa√m)	Censored (Y/N)	Censored 1T-K _{Jc} value (MPa√m)	T₀Q (°C)	ΔT₀₀ (°C)
N000L	-139	103.6	62.2	Υ	59.8		
N000LM	-140	55.8	38.1	Υ	59.8		
N000RM	-140	67.4	43.9	Ν	NA		
N000R	-140	45.2	32.7	Ν	NA	71	21
N001L	-136	176.8	99.1	Ν	NA	-/ 1	21
N001LM	-140	23.9	22.0	Υ	61.9		
N001RM	-140	35.8	28.0	Ν	NA		
N001R	-140	75.2	47.9	Ν	NA		

Table 11. Fracture toughness of irradiated Eurofer97 material O

Notch ID	Test temperature (°C)	K _{Jc} (MPa√m)	1T-K _{Jc} (MPa√m)	Censored (Y/N)	Censored 1T-K _{Jc} value (MPa√m)	T₀Q (°C)	ΔΤ _{οQ} (°C)
O000L	-105	34.7	27.4	N	NA		
O000LM	-110	38.7	29.5	N	NA		
0000RM	-101	129.5	75.3	Y	66.8		
0000RM	-98	51.7	36.0	N	NA	40	20
O001L	-100	29.7	24.9	N	NA	-40	30
O001LM	-99	211.9	116.9	Y	66.2		
0001RM	-102	46.0	33.1	N	NA		
0001R	-100	43.7	32.0	N	NA		

Notch ID	Test temperature (°C)	K _{Jc} (MPa√m)	1T-K _{Jc} (MPa√m)	Censored (Y/N)	Censored 1T-K _{Jc} value (MPa√m)	T₀Q (°C)	ΔΤ _{οQ} (°C)
P000L	-100	289.0	155.8	Υ	63.8		
P000LM	-106	38.2	29.2	Ν	NA		
P000RM	-104	25.8	22.9	Ν	NA		
P000R	-98	82.1	51.3	Ν	NA	40	16
P001L	-98	226.8	124.4	Υ	64.4	-42	40
P001LM	-100	93.4	57.1	Ν	NA		
P001RM	-99	70.0	45.2	Ν	NA		
P001R	-99	40.3	30.3	N	NA		

Table	12.	Fracture	toughness	of irradiated	Eurofer97	material P
					-	

Lastly, the correlation between the upper shift in T_{0Q} and change in Vickers microhardness for ten Eurofer97 variants after irradiation is shown in Figure 7. Materials M and N showed minimum irradiation hardening and embrittlement due to the elevated irradiation temperature. A linear correlation between the upper shift in T_{0Q} and change in Vickers microhardness was observed for only a few materials indicating different irradiation response of the Eurofer97 variants. Indeed, based on the report by Gaganidze [9], standard Eurofer97 is expected to show an upper shift of 130°C in T_0 after irradiation at 300°C to 2.5 dpa. Materials E, K, and L showed a similar degree of irradiation embrittlement as the standard Eurofer97 whereas Materials H, I, J, O and P showed less embrittlement. Further microstructure study is needed to understand the difference in the observed fracture toughness results.





Conclusions

In this study, we performed the post-irradiation evaluation of ten Eurofer97 variants after the nominal HFIR irradiation at 300°C to 2.5 dpa. Main findings of this study include:

1) We have successfully developed a small specimen testing technique for fracture toughness testing. The technique utilizes pre-cracked miniature multi-notch bend bar specimens (M4CVN) with testing and analysis based on the Master Curve method in the ASTM E1921 standard.

2) Two Eurofer97 variants (materials M and N) experienced a much higher irradiation temperature of ~476°C which resulted in negligible irradiation embrittlement and hardening.

3) The remaining Eurofer97 variants (materials E, H, I, J, K, L, O, and P) reached essentially target irradiation conditions and showed different degrees of irradiation embrittlement and hardening.

4) For the same irradiation condition, materials E, K, and L showed a similar degree of irradiation embrittlement as the standard Eurofer97 whereas Materials H, I, J, O and P showed less embrittlement.
5) A linear correlation between the upper shift in T_{0Q} and change in Vickers microhardness was observed for only a few Eurofer97 variants indicating different irradiation response of those materials.

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1.3 PERFORMANCE IMPROVEMENT OF MODIFIED 3Cr BAINITIC STEEL WELDMENTS—Y. Yamamoto (Oak Ridge National Laboratory)

OBJECTIVE

This work aims to develop new bainitic steels, based on 3Cr-3WV(Ta) steels originally developed at ORNL. The goal is mechanical properties of both base metal and weldments superior to those of existing commercial bainitic steels or ferritic-martensitic (F-M) steels, together with no requirement for post-weld heat treatment (PWHT). The target applications are high-temperature structural components in fusion reactors such as vacuum vessel, structural ring which supports the blanket modules, and magnet shields, to be used at or above the 400-500°C range. Improvements of creep performance and room-temperature toughness are targeted via optimization of alloy composition and thermo-mechanical treatment.

SUMMARY

Detailed microstructure characterization of the creep-ruptured 3Cr-3WV base bainitic steel weldments were conducted. Modified 3Cr-3WV base steels, MLC02 and MLC02B (high Mn + low C without and with 0.01 B, respectively) showed a slightly longer and shorter cross-weld rupture-lives, respectively, than that of the original 3Cr-3WV steel weldment at 600°C and 170MPa. The original 3Cr-3WV steel and MLC02 showed a large deformation concentration, as well as creep-rupture, at the inter-critical heat affected zone (ICHAZ), which was attributed to microstructure instability at the ICHAZ during creep testing. MLC02 showed slightly better microstructural stability than the original, which could result in improvement of creep-life. On the other hand, MLC02B exhibited further improved microstructural stability as well as no significant deformation concentration at the ICHAZ. However, less deformability caused unexpectedly short rupture-life compared to the others. It was found that the improvement of microstructural stability at the ICHAZ would be the key for the cross-weld creep performance, and the B addition is not the direct solution of the creep property improvement. The 3Cr-3WV base steel with the Ta addition is currently considered for further evaluation.

PROGRESS AND STATUS

Based on previous results, two new 3Cr-3WV base bainitic steels (MLC02 and MLC02B, in Table 1) were prepared which incorporated the proposed design strategy with the additions of high Mn, low C, low Si, and with/without a small amount of B. The boron addition in ferritic-martensitic steels is known to improve the thermal stability of the carbide which supports stabilization of the martensite laths at elevated temperatures, and therefore the creep performance as well [1]. Helium-bubble formation due to transmutation of B in the service condition is sufficiently small or negligible because of relatively low expected doses in the target components compared to the others. In addition, the new alloys were based on Ta-free 3Cr-3WV steel in order to eliminate the combined effect of MC type carbide formation on the alloy modification. In this report, the cross-sectional microstructure of the cross-weld creep tested specimens has been characterized to understand the microstructure degradation during creep deformation.

Alley	Nomir	nal com	Demerika							
Alloy	Fe	Cr	Mn	Si	Та	V	W	С	В	Remarks
MLC02	91.59	3	2	0.16	-	0.2	3	0.05	-	High Mn, Iow C
MLC02B	91.58	3	2	0.16	-	0.2	3	0.05	0.01	High Mn, Iow C, with B
Original	93.14	3	0.4	0.16	-	0.2	3	0.1	-	3Cr-3WV base, Ta-free

Table 1. Nominal and analyzed compositions of the steels

Microstructure characterization indicated that the inter-critical heat affected zone (ICHAZ) was the key to control the creep-performance of the weldments. The cross-weld creep-rupture tests of three steels were conducted at 600°C in a laboratory air and various stresses from 130 to 250 MPa (constant loading). The rupture-lives of the steels at 600°C and 170MPa were comparable to each other, although MLC02 showed slightly longer rupture-life than the original (241h and 184h, respectively). MLC02B exhibited the shortest rupture-life (156h) among three steels. Figure 1 represents the cross-sectional macrostructure of the creep-ruptured specimens tested at 600°C and 170MPa. The original 3Cr-3WV steel showed a large deformation, as well as the creep-rupture, at the ICHAZ which locates just outside of the fine grain heat affected zone (FGHAZ). Similar deformation concentration at the ICHAZ was observed in MLC02. MLC02B also ruptured at the ICHAZ like the others.



Figure 1. Cross-sectional view of cross-weld creep-ruptured specimens tested at 600°C and 170MPa (Pretested specimens: N+T+GTAW, no PWHT). The yellow lines correspond to the areas where the hardness measurement was conducted.

The hardness measurement across the HAZ indicated that a significant hardness drop occurred at the ICHAZ during creep testing. Figure 2 illustrates the color contour map showing the hardness distribution inside the creep-ruptured specimens tested at 600°C and 170MPa. All steels showed the minimum hardness at the ICHAZ, which was even lower than that of the base metal, indicating that the deformation concentration during creep testing could strongly be correlated with the hardness drop at the ICHAZ. It should be noted that the hardness drop in MLC02B at the ICHAZ was not so significant compared to the others, despite the least rupture-life among three steels, suggesting that some other mechanisms deteriorating the creep performance at the ICHAZ also need to be considered.



Figure 2. 2D color contour maps illustrating the hardness distribution across the weld metals in the cross-weld creep-ruptured specimens shown in Figure 1.

Comparison of the hardness distribution between the as-GTAW and creep-ruptured specimens is also shown in Figure 3. As previously reported, the variation of the hardness in the as-GTAW material was large in the original 3Cr-3WV steel, whereas those in MLC02 and MLC02B were relatively small. After creep testing, the hardness inside the HAZ in the original 3Cr-3WV steel showed less variation, and the hardness seemed quite low. MLC02 exhibited some hardness variation inside the HAZ with minimum hardness at the ICHAZ. The hardness inside the HAZ was slightly higher than that in the original 3Cr-3WV steel. These comparison suggests that the cross-weld creep performance could be improved by increasing the thermal stability of the microstructure at the ICHAZ. The MLC02B, on the other hand, showed relatively high hardness compared to the others with similar hardness distribution before and after testing, indicating that the microstructural stability was improved by the B addition. However, it also showed the short rupture-life among three steels. This might be attributed to less deformability of MLC02B, due to the B addition, which gave another detrimental effect on the cross-weld creep-rupture performance.



Figure 3. Comparison of the averaged hardness distribution across the heat-affected zones before and after cross-weld creep-rupture testing; (a) original, (b) MLC02, and (c) MLC02B.

Based on the results, it was found that the improvement of microstructural stability at the ICHAZ would be the key for the cross-weld creep performance. However, the B addition was not a direct solution to improve the performance despite the improved microstructural stability. The Ta addition is considered for improvement of the microstructural stability and the cross-weld performance as well. A new heat production with the Ta addition for further evaluation is currently in progress.

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1.4 BUBBLE FORMATION IN HELIUM-IMPLANTED NANOSTRUCTURED FERRITIC ALLOYS AT 500°C and 700°C—Yan-Ru Lin, David T. Hoelzer, Lizhen Tan (Oak Ridge National Laboratory), Steven J. Zinkle (University of Tennessee)

OBJECTIVE

The objective of this task is to study the cavity size and density in several He-implanted nanostructured ferritic/martensitic alloys (CNA1, CNA3, 9YWTV and 14YWT) with different starting dispersoid sink strengths. The synergistic effect of He along with radiation damage can cause unwanted degradation of the mechanical performance of structural materials and impact the safety of fusion power plants. It has been proposed that these effects could be mitigated by increasing the number of He trapping sites to control the bubble size or to shield He from the grain boundaries. This concept has led to the development of high sink strength materials with nanoclusters, such as Oxide dispersion strengthened (ODS) alloys or alloys with MX type precipitates. However, there is no systematic irradiation data showing how the density of nanoclusters and their He trapping ability (binding energy) in advanced ferritic alloys affect the helium bubble density and size at elevated temperatures.

SUMMARY

Cavity formation in Fe-9Cr and nanostructured ferritic/martensitic alloys CNA1, CNA3, 9YWTV, and 14YWT after He implantation (to ~7000 appm at 500°C and 700°C) was examined by transmission electron microscopy (TEM). From 500-700°C, the bubble size was observed to increase with increasing irradiation temperature for all the materials. In the same temperature range, the bubble density decreased with increasing temperature. At both temperatures, the cavity size in the implanted materials was in the order of Fe-9Cr > CNA1 > CNA3 >9YWTV>14YWT, while the cavity density showed the opposite order. In addition, the observed bubble number densities for the four nanostructured alloys are comparable to the nanocluster density, suggesting that the nanoclusters in both alloys were very effective in trapping He at elevated temperatures. The results indicated that the 14YWT alloy appears to sequester the helium into smaller bubbles more effectively than other alloys, which led to a lower volume swelling value. This can be attributed to the much higher sink strength associated with the nanoclusters in the 14YWT alloy. This conclusion agrees with the results of our in-situ TEM study on He implanted CNA3 and 14YWT materials.

PROGRESS AND STATUS

In our previous progress report [1], He bubbles were only found in the He implanted Ni material, but not in the 14YWT and CNA3 alloys at 500°C and 700°C. However, after improving the FIB sample quality with thinner foil thickness (~50nm) and lower FIB surface damage (by using a low voltage FIB probe longer during the final polishing process), cavities were visible in both nanostructured alloys at 500°C and 700°C. In another previous progress report [2], we performed in-situ helium implantation on TEM samples of a high purity Fe-10Cr alloy and two other nanostructured ferritic/martensitic (FM) alloys (14YWT and CNA3) utilizing the IVEM facility at Argonne national laboratory.

In a follow-up study on bulk irradiated materials, 275 keV He ions were used to irradiate a model Fe-9Cr alloy, and four different nanostructured FM alloys (9YWTV, 14YWT, CNA1 and CNA3) [3][4] at 500°C and 700°C with a fluence of 1.28 x 10^{20} m⁻². Based on the SRIM calculation, the peak radiation damage (at ~700 nm) and He concentration were roughly 0.3 dpa and 7000 appm, respectively. The general microstructures of the five materials following He implantation at 500°C and 700°C are shown in Figure 1 and Figure 2, respectively. Higher magnification TEM examination showed that the size of the cavities (He bubbles) increased with increasing irradiation temperature, while the bubble density decreased with increasing temperature. Furthermore, the cavities were mostly uniformly distributed in the range of 500-800nm from the implanted surface. The peak damage depth is indicated in red dashed lines in both Figure

1 and Figure 2. However, for the 700°C He-implanted Fe-9Cr model alloy (i.e., without the addition of nanoclusters), bubbles were agglomerated along the primitive dislocations showing a heterogeneous distribution (Figure 2). This is evidence that He bubble precipitation can be more heterogeneous at high temperatures, due to higher mobility of small He-vacancy clusters compared with lower temperatures.



Figure 1. Low-magnification TEM images of cavities distribution of He implanted materials at 500°C.



Figure 2. Low-magnification TEM images of cavities distribution of He implanted materials at 700°C.

In order to quantify the cavity size and density, higher magnification TEM images of all the samples were taken near the depth of 700 nm, as shown in Figure 3. Further, the cavity size, density, and volume swelling measurements are summarized in Table 1. At both 500°C and 700°C, Fe-9Cr (no nanoclusters) had the largest diameter (3.71 and 8.75 nm, respectively) but lowest density (1.05x10²³m⁻³ and 1.95 x10²²m⁻³, respectively). Comparing the model Fe-9Cr alloy with the nanostructured materials, the cavity size was in the order of Fe-9Cr > CNA1 > CNA3 >9YWTV>14YWT, while the cavity density showed exactly the opposite sequencing. Considering the temperature effect, the size increment and density decrement also has the same order, i.e. for a given implanted material the smallest size and largest density occurred at the lower temperature (500°C). Looking at the two materials with the lowest and highest sink strength, with increasing temperature the cavity sizes in Fe-9Cr and 14YWT increased about 135% and 1%, respectively. For the density dependence on temperature, Fe-9Cr and 14YWT had 81% and 26% decrement, respectively. Lastly, cavity volumetric swelling was estimated by calculating the volume of cavities with respect to the volume of the analyzed zone assuming that each cavity is spherical. As noted in Table 1, 14YWT alloy also showed the lowest total volume swelling, while Fe-9Cr had the largest swelling value. The temperature dependence of the cavity diameter and density of the bulk irradiation (filled symbol and solid line) and previous in-situ irradiation results (unfilled symbol and dashed line) are illustrated in Figure 4.



100 nm

Figure 3. Cavities in He implanted materials

		madiated material	5		
Temperature(°C)	Materials	Average Size (nm)	Volume Density(10 ²² m ⁻³)	Swelling (%)	
	Fe-9Cr	3.71	10.47	0.59	
	CNA 1	2.54	11.83	0.13	
500	CNA 3	2.24	19.34	0.12	
	9 YWTV	2.08	26.42	0.13	
	14YWT	1.82	38.10	0.09	
	Fe-9Cr	8.75	1.95	1.20	
	CNA 1	4.62	3.48	0.31	
700	CNA 3	4.28	3.56	0.21	
	9 YWTV	2.24	22.5	0.14	
	14YWT	1.84	27.86	0.13	

Table 1. Quantitative result on average diameter, number density, and volume swelling of cavities in irradiated materials





In summary, from 500°C to 700°C, the general trend showed that bubble size increased, and the density decreased for all alloys, as expected based on simple cavity coarsening expectations and numerous prior studies on gas-implanted materials [6]. However, the variation was much more moderate for the 14YWT alloy that had the highest nanocluster/precipitate density. According to previous APT and TEM studies [3-5, 7], the observed bubble number densities summarized in Table 1 are comparable to the nanocluster densities for CNA3 and 14YWT (N_{He}/N_c ~1), suggesting that the nanoclusters in both alloys were effective at trapping He at elevated temperatures. Conversely, the two CNA alloys exhibited good He trapping at 500°C but relatively inefficient trapping at 700°C, based on the dramatic decrease in observed cavity density in these two alloys at 700°C (Table 1). The results indicate that the 14YWT alloy appears to sequester the helium into smaller bubbles more effectively than other alloys. This may be attributed to the much higher

sink strength associated with the nanoclusters in the 14YWT alloy, or the difference of the He trapping ability between oxides and MX precipitates.

Future Plans

According to the previous APT and TEM studies of the nanocluster density in CNA (with MX type particles) and 9/14-YWT (with Y/Ti oxides) alloys, the particle density follows the order of 14YWT>9YWTV>CNA3>CNA1, which is as same as the order of cavity density in the He implanted materials in this study. A detailed APT, EELS or EDS study on the size and density of the nanoclusters in the materials used in this study in planned for the next step to understand the detailed spatial relationship between the nanoclusters and the cavities (He bubbles).

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1.5 A FUSION RELEVANT ENGINEERING VOID SWELLING MODEL FOR 9Cr TEMPERED MARTENSITIC STEELS—Takuva Yamamoto, G. Robert Odette (University of California Santa Barbara)

OBJECTIVE

The objective of this research is to develop an engineering model for void swelling (f_v) in 9Cr tempered martensitic steels (TMS) as a function of dpa for fusion service relevant effects of the displacements per atom (dpa), dpa rate (dpa'), helium (He) and the helium to dpa ratio (He/dpa).

SUMMARY

The UCSB database on cavity evolution, including the results of both dual ion irradiation (DII) and HFIR in situ helium injection (ISHI) neutron irradiation experiments, were combined with data from the literature to develop a f_v(dpa, dpa', He/dpa) model for 500°C (400-500°C including literature data) irradiations of 9Cr TMS. The TMS cavity evolution 500°C DII observations include 346 material-dpa-He-dpa' data sets. The corresponding 500°C neutron ISHI irradiation observations include 16 material-dpa-He-dpa' data sets. The UCSB database was complemented by many data sets on similar single ion irradiations (SII) and neutron irradiations (NI) of TMS found in the literature. In the case of the DII irradiations, f_v(dpa, dpa', He/dpa) is strongly affected by the He/dpa ratio, particularly with respect to the incubation dpa (dpai) for the onset of void swelling. However, He/dpa also affects the post-incubation swelling rate (f_v). The large "scatter" in the DII irradiation f_v is primarily the result of local microstructural variations and is mainly due to local microstructural effects on injected interstitial (II) atoms in single ion irradiations (SII) and DII. Local microstructures that shield regions at shallower depths from II, provide the most accurate measures of dpaⁱ and f_v ; and, more generally, the highest f_v is taken the best measure of void swelling rates. The incubation dpaj is lower in the ISHI NI. SII and NI, resulting in low He/dpa ratios, require a much larger dpai, but which is again less in NI. The ion versus neutron irradiation differences are likely due to the much lower dpa' in the latter case. The combined data set suggests that, after the initial incubation transient, f_v can reach up to $\approx 0.3\%$ /dpa (see below), which is even higher than the canonical rate of 0.2%/dpa proposed by Garner [1]. This f_v is also higher than the \approx 0.1%/dpa found in the UCSB DII database, probably due to the lower post-incubation DII dpa increment, which is still in the transient regime. The post incubation fv' can be qualitatively rationalized in terms of the void, bubble and dislocation sink ratios. A master model of cavity evolution in TMS is also semi-qualitatively consistent with the experimental results and provides additional mechanistic insight. The best-fit He/dpa = 10 (in units of appm He/dpa) model predicts swelling of 0.8, 3.3, 6.7 and 12.4% at 50, 100, 150 and 200 dpa, respectively, with estimated uncertainties of ±50%.

Background

Fusion reactor designs must account for dimensional instabilities in structural components associated with void swelling and irradiation creep. It is well established that high He and He/dpa levels can cause severe degradation of both mechanical properties and acceleration of void swelling in TMS [2]. Thus, irradiation damage in a He-rich environment may greatly narrow the application window for the current candidate TMS for fusion reactor first wall and blanket structures. Here we analyze the DII, ISHI and literature SII and NI databases to develop a predictive swelling model.

PROGRESS AND STATUS

Introduction

UCSB TMS swelling database

The UCSB has been developing microstructure database based on two types of simultaneous dpa damage and He implantation irradiations. One is the in-situ He injection (ISHI) method in mixed spectrum fission reactor irradiations using Ni (or B or Li)-bearing implanter layers to inject high-energy α-particles into an adjacent material that is simultaneously undergoing fast neutron induced displacement damage. as illustrated in Figure 1a [2-4]. A series of in-situ He implantation irradiation experiments have been carried out in the mixed spectrum High Flux Isotope Reactor (HFIR) [3-9]. Micron scale NiAl injector coatings were used to uniformly implant α -particles to a depth of ≈ 5 to 8 µm in transmission electron microscopy (TEM) discs for a large matrix of alloys irradiated over a wide range of temperatures and dpa at controlled He/dpa ratios ranging from << 1 to 65 appm/dpa. The other approach is to use dual ion beam irradiations (DII) to simultaneously implant He ions and create displacement damage with heavy ions [10-13]. The DII were performed in DuET facility at the Institute of Advanced Energy, Kyoto University in Japan. Here, Fe³⁺ and He⁺ ions are accelerated to 6.4MeV and 1MeV, respectively, to create dpa damage and He profiles as illustrated in Figure 1b. The He⁺ ion beam is passed through a rotating energy degrader foil with carrying thicknesses to create four ion energy ranges, and corresponding implantation depth regions, that result in a broader and more uniform He/dpa deposition profile up to a maximum depth of \approx 1500 nm. The ion beams are rastered in DuET.

Note, the two techniques are complementary but manifest many differences that, in the case of DII. include: a) much higher dpa rates; b) non-uniform spatial distributions of dpa and He; c) the proximity of a free surface; and d) the Fe ions deposited as injected self-intestinal atoms (II) near the dpa peak region. Figure 1c shows that the spatial dpa-He distribution can probe many combinations of He-dpa in a single experiment, while Figure 1d compares dpa-dpa rate combinations in DII to that in ISHI reactor irradiations as well as in spallation proton (STIP) irradiations.



appm

He

Figure 1. a) Schematics of in situ He injection experiments; b) an example of dpa, He and He/dpa profiles in DuET experiments; c) the dpa-He; and, d) dpa-dpa' conditions in UCSB irradiation experiments using various techniques, including DII DuET at discrete depth increments where quantitative TEM was carried out.

The DII Database

We have updated our DII microstructure database with newly analyzed cavity microstructural data for F82H Mod.3. To date we have collected data on 346 irradiation (dpa, He, dpa rate)-material condition combinations, including 136 conditions for F82H Mod.3 and 98 conditions for F82H IEA both in astempered (AT) materials as well as 112 conditions for cold-worked (CW) F82H IEA. Table 1 summarizes nominal conditions (at two depth locations) for the DII experiments carried out at 500°C.

Evo ID	T (°C)	Nominal Condition (@550-650nm)			Peak He (@1000-1100nm)			Allovs	
	dpa	(appm)	He/dpa	dpa/s	dpa	(appm)	He/dpa	Alloys	
DI10B1	500	26	1210	47	5.0 x 10 ⁻⁴	45	2100	47	F82H mod.3, MA957
DI10B2	500	9.9	457	46	5.2 x 10 ⁻⁴	17	795	46	F82H mod.3, MA957
DI10B3	500	10	480	47	5.1 x 10 ⁻⁴	18	840	47	F82H mod.3, MA957
DI13A	500	26	390	15	5.1 x 10 ⁻⁴	44	670	15	mod.3, IEA (AT,20,80), MA957
DI13B1	500	30	848	29	1.5 x 10 ⁻³	51	1467	29	F82H mod.3, F82H IEA (AT)
DI13B2	500	27	730	27	1.6 x 10 ⁻³	46	1262	27	F82H IEA (20%CW or 80%CW)
DI13B1+2	500	57	1578	28	1.5 x 10 ⁻³	97	2729	28	MA957
DI14A1	500	30	1200	47	1.3 x 10 ⁻³	45	2100	47	F82H mod.3, F82H IEA (AT)
DI14A2	500	30	1200	47	1.3 x 10 ⁻³	45	2100	47	F82H IEA (20%CW or 80%CW)
DI14A1+2	500	52	2400	47	1.3 x 10 ⁻³	90	4200	47	MA957
DI14B	500	45	1290	28	8 x 10 ⁻⁴	79	2230	28	F82H mod.3, F82H IEA (AT)
DI15A1	500	51	1360	27	8.6 x 10 ⁻⁴	88	2350	27	F82H mod.3, IEA (AT,20,80)
DI16A	500	51.5	2327	45	6.5 x 10 ⁻⁴	89.2	4024	45	F82H mod.3, IEA (AT,20,80)
DI16A+19A	500	81.5	3748	45	6.27 x 10 ⁻⁴	141.7	6397	45	F82H mod.3, IEA (AT,20,80)

Table 1.	The nomina	l dpa-He	conditions	of DII	experiments
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Figure 2 shows an example of the microstructure evolutions in DII experiments over the entire depth range (top) from the surface (left), where the dpa peaks at $\approx 1.6\mu$ m, tailing off at $\approx 2\mu$ m. The He concentration peaks at $\approx 1.1\mu$ m tailing off $\approx 1.5\mu$ m. Higher magnification micrographs compare the cavity microstructures at 25 and 45 dpa, for He/dpa ≈ 47 , and at He/dpa ≈ 15 and 47 for 45 dpa.



Figure 2. An example of cavity (bubbles and voids) evolution over the depth from ion irradiation surface (top); and higher magnification micrographs comparing 25 versus 45 dpa at He/dpa \approx 46 appm/dpa, as well 15 versus 45 appm He/dpa at 45 dpa.

Figure 3 summarizes dpa and He/dpa ratio dependence of the cavity microstructures in F82H IEA in terms of: a) the number density (N_v); b) the average diameter (<d_v>); and, c) the volume fraction (f_v) of "voids", defined as cavities larger than 4 nm. While the data are highly scattered, the eyeball lines define broad trend bands at He/dpa > 35, \approx 30 and \approx 20, and suggest that N_v, <d_v> and f_v increase monotonously with both dpa and He/dpa.



Figure 3. a) The number density (N_v) ; b) average diameter (<d_v>); and, c) volume fraction (f_v) of "voids" in F82H IEA as a function of dpa and the He/dpa ratio.

The ISHI database

The ISHI microstructure database includes 13 irradiation (dpa and He)-material conditions (10 for AT and 3 for CW materials) at 500°C. Figure 4a shows evolution of the cavity microstructure in a TMS Eurofer 97 steel from the lowest 9 dpa-190 appm He condition (top left) to higher He/dpa (bottom left), or higher dpa (top right), or higher appm He (bottom right). The ISHI data clearly indicate that higher He/dpa accelerates void nucleation and higher dpa increases f_v at a specified He/dpa. Figure 4b plots f_v as a function of dpa, an increasing swelling rate f_v '.



Figure 4. a) The ISHI cavity microstructure in a TMS Eurofer 97 steel from 9 dpa-190 appm He (top left) to higher He/dpa (bottom left) or higher dpa (top right) up to the highest 21 dpa, 1200 appm He condition (bottom right); and, b) the corresponding f_v as a function of dpa.

The literature database

We also have collected swelling data on TMS ferritic steels and Fe-Cr model alloys from the literature [1,14–29]. This database currently consists of f_v (and usually N_v and <d_v>) for 95 NI and 83 SII data sets for F82H, 9Cr-Mo (T91) and HT9 TMS as well as Fe-3-15Cr binary model alloys. Analysis of the literature database leads to the following general observations:

- Ferritic materials reach swelling rates from f_v = 0.1 to 0.3%/dpa.
- Injected interstitials (II) have strong effect in suppressing swelling in single ion irradiation (SII) experiments.
- The incubation dpa, for ions is higher for SII compared to NI.
- Pre-injecting with He followed by SII is not a good simulation of He effects.

Injected SIA (II) effects on swelling

A significant and growing literature demonstrates the profound effect of II on void swelling in ion irradiations [17, 26-29]. This is due to the fact the void swelling is driven by small difference between vacancy and interstitial fluxes that control the critical bubble to void conversion radius (r_c) and the incubation dpa (dpa_i) as well as the net f_v ' in the post incubation regime. In simple rate theories of swelling, the excess vacancy flux to voids is due to the bias of dislocations for SIA. Thus, the addition of even small concentrations of II can have a huge impact on the net excess vacancy flux to voids. Hence, II reduce swelling rates and increase the critical radius needed to convert stable bubbles to growing voids. Since SIA diffuse rapidly, the II effect may extend beyond the ion deposition end of range deposition profile. Most data suggest the II decreases significantly by about 500 ± 250 nm from the dpa peak. Swelling is also reduced close to the implanted surface. Thus, there is only a limited region to characterize f_v and f_v '.

However, the presence of a grain boundary can block the diffusion of II towards the specimen surface. This is illustrated in Figure 5 for F82H Mod.3, where a grain boundary decorated with small He bubbles shields the adjoining region nearer the surface, which contains large voids resulting in a significant f_v is observed. The dpa in this region increases approximately linearly from 53 to 86 dpa between 0.15 and 0.65 μ m. The He/dpa increases from \approx 13 at 0.15 μ m reaching a roughly constant 50 appm/dpa between 0.35 μ m and 0.7 μ m. The corresponding f_v also increases approximately linearly with depth in this region, up to \approx 4.5%, as indicated by the black dashed line. A fit to the f_v -dpa data, yields a slope of 0.12 %/dpa. The intercept is dpa_i \approx 53 and 13 appm He/dpa; note, however, that the intercept depth is in fairly close proximity to the surface.

Figure 6 shows another example of higher f_v in the shadow of a boundary at an $\approx 1.5 \mu m$ depth (73 to 82 dpa at ≈ 10 appm He/dpa on average) and a group of larger carbides at an $\approx 1.1 \mu m$ depth (30 to 48 dpa at ≈ 30 appm He/dpa). Similar features were found in almost every case that showed high swelling. Assuming such local microstructural features blocking II diffusion are the main cause of high swelling, among highly scattered data, we take the highest f_v ' data as being closest to the unaffected swelling rate. This concept is used to analyze the DII database, and in combination with ISHI and literature data, to derive a predictive engineering $f_v((dpa, dpa', He/dpa)$ swelling model for a fusion relevant He/dpa ≈ 10 appm/dpa at $\approx 500^{\circ}$ C. Note, this is also the best current basis to estimate f_v for lower temperatures down to $\approx 400^{\circ}$ C.

Figure 7 plots the highest f_v versus dpa data in groups of He/dpa of > 40, 30, 20, and 10. The f_v for He/dpa < 5 has been excluded because of the very narrow dpa increment of post-incubation f_v data. Least square fits to the highest f_v data were carried out in two ways: a) f_v as a function of dpa; and, b) dpa as a function of f_v . The two procedures result is in slightly different dpaⁱ and f_v '. Figure 8 plots dpa_i and f_v ' as a function of He/dpa. Both decrease systematically with increasing He/dpa. The decrease of dpa_i higher He/dpa, is due to reaching critical bubble sizes at lower dpa. The reduction in f_v ' is likely due to the increase in the bubble sink strength with increasing He/dpa.



Figure 5. a) Cavity formation (f_v) – depth trends a DII F82H Mod.3 sample along with the dpa and He conditions; and, b) cavity image of the area showing high f_v behind a boundary at a depth $\approx 0.8 \mu$ m.



Figure 6. An example of high f_v regions that are shielded from the II in the shadows of a grain boundary and some carbides.



Figure 7. The high f_v data as a function of dpa fitted as: a) $f_v(dpa)$; and, b) dpa(f_v).

Figure 8a plots the dpa_i versus He/dpa. The fit line to the DII data decreases with He/dpa that is represented by a hockey stick form. There is only one reliable data point at a He/dpa \approx 46 appm/dpa for the ISHI condition. The linear trend in the DII data suggests that there is a threshold He for the onset of void swelling. The average He_i = [dpai]x[He/dpa] data \approx 890 ± 200 appm. The value for the fitted DII curve is He_i = 769±77. The corresponding He_i for the ISHI irradiation is 325 appm, or a dpa_i \approx 33 at He/dpa \approx 10 appm/dpa. Figure 8b plots f_v' as a function of He/dpa. Although the f_v data are highly scattered, the fitted trend line shows that f_v' decreases with increasing He/dpa. Again, this trend is likely due to a corresponding increase in the bubble sink strength at higher He/dpa ratios.



Figure 8. a) The dpa_i versus He/dpa for the two ways of fitting, along with the solid fit line and the H_{ei} scaled dpa_i dashed line for the ISHI; and, b) the post incubation f_v ' as a function of He/dpa.

Figure 9a plots the fitted DII post-incubation f_v for He/dpa groups from 10 to > 40 He/dpa. The f_v ' is lower for He/dpa \approx 30 appm/dpa but increases at lower He/dpa of 20 and 10 appm/dpa. Figure 9b shows f_v versus dpa normalized to a common He/dpa = 10 appm/dpa based on the relations shown in Figure 8a and b: a) dpa_n = dpa - dpa_i(He/dpa) + dpa_i(10); and, b) $f_{vn} = (dpa - dpa_i)^* \{f_v' + 0.00168 (He/dpa - 10)\}$.

The normalized f_{vn} fall in a narrow band with an overall post-incubation swelling rate ≈ 0.115 %/dpa and an incubation dpa_i $\approx 72-78$ dpa.



Figure 9. a) f_v as a function of dpa - dpa_i for various He/dpa ratios; and, b) post-incubation f_{vn} data normalized to a common dpa_i and f_v for He/dpa = 10 appm/dpa.

Figure 10a plots the sinks strength ($Z_i \approx 4\pi r_i N_i$) of bubbles and voids as a function of f_v . The void sink strength (Z_v) first increases with f_v and then levels off; the bubble sink strength (Z_b) decreases with f_v , due to conversion to voids. Along with SIA biased (B) dislocations sinks (Z_d), void and bubble Z_v and Z_b control the post-incubation f_v ', if Gibbs Thomson effect vacancy emission from cavities is assumed to be small, as:

$$f_{v} = \eta B Z_{d} Z_{v} / [(Z_{b} + Z_{v} + Z_{d})(Z_{d}(1 + B) + Z_{v} + Z_{b})]$$
(1)

Here $\eta \approx 0.33$ is the vacancy and SIA generation rate per dpa. Figure 10b shows that this simple sink partitioning swelling model is consistent with the observed trends, and only slightly over predicts the f_v data, likely due to vacancy emission from voids that is not accounted for.



Figure 10. a) Z_v and Z_b as a function of f_v ; and, b) the f_v ' predicted by Equation 1 as a function of Z_v/Z_d for various Z_b/Z_v for $Z_d = 10^{15}/m^2$ and B = 0.015.

<u>Blending the DII, ISHI and literature low He neutron and SII data to derive a fusion relevant f_v(dpa. He/dpa) model</u>

Figure 11a shows f_v as a function of dpa for low He NI and SII literature data. Both f_v (dpa) data sets can be fit with the same empirical fourth order polynomial,

 $f_{v} = 1.27 \times 10^{-2} (dpa-dpa_{i})^{+} 1.03 \times 10^{-3} (dpa-dpa_{i})^{2} - 7.26 \times 10^{-6} (dpa-dpa_{i})^{3} + 2.63 \times 10^{-8} (dpa-dpa_{i})^{4}$ (2)

Equation 2 is applicable to dpa > dpa_i, which are \approx 379 dpa for low He SII and \approx 159 dpa for low He neutron irradiations. Figure 11b shows the corresponding collapsed f_v plots versus dpa - dpa_i with an increasing f_v up to > 0.3%/dpa.



Figure 11. a) Literature f_v as a function of dpa for low He SII and neutron irradiations, fitted with a fourth order polynomial; and, b) the corresponding collapsed literature f_v as a function of dpa - dpa_i, along with the same fit curve.

Figure 12a shows the same polynomial f_v versus dpa - dpa_i curve including the DII and ISHI NI data. Here the DII dpa_i is normalized to 50 dpa at a He/dpa = 10. Note this dpa_i is lower than in Figure 12 due the polynomial curvature of f_v (dpa-dpa_i). The corresponding ISHI data at He/dpa \approx 22 and 50 appm/dpa fitted to the literature f_v (dpa-dpa_i) polynomial curve give dpa_i = 11 and 4.4, respectively, based on a common nominal threshold He_i \approx 220 appm. Figure 12b shows the f_v prediction for He/dpa=10, along with an estimated uncertainty band of ± 50%.

Discussion

The empirical $f_v(dpa, He/dpa)$ is based on use and synthesis of essentially all the data that is available on void swelling in TMS. The high f_v and dpa data is almost entirely based on low He, high dpa' SII results that may not be representative of fusion relevant neutron irradiations. A full and detailed discussion of the issues that ion versus neutron irradiations raise is beyond the scope of this initial report. However, we believe that empirically accounting for the effect of He/dpa on dpa_i and f_v ' at 500°C and the corresponding effects of low versus high dpa' on dpa_i, at both low and high He/dpa, is a reasonable basis to derive a fitted model of $f_v(dpa, He/dpa)$ for fusion relevant conditions. However, higher dpa ISHI and DII data are clearly urgently needed to further refine the swelling model. A roadmap for acquiring such data has been developed, but further discussion of these opportunities is also beyond the scope of this report.
However, some additional insight is provided by predictions of a detailed cluster dynamics, rate theory He generation, transport, fate and consequences (GTF&C) master model, applied here to void swelling [30]. Figure 13a shows the model predictions compared to the ISHI data. The solid red curve is for 45 appm He/dpa, while the dashed green curve is for 15 appm He/dpa. The data points are for 20 to 60 appm He/dpa. The higher He/dpa curve bends over due to the buildup of bubble sinks. The lower He/dpa curve is linear and shows a dpa_i \approx 12 and post incubation f_v' \approx 0.05%/dpa. These are both consistent with the ISHI data. Figure 13b show the corresponding model predictions for the DII f_v data. The dpa_i are significantly under predicted by the model, which also indicates a smaller effect of He/dpa compared to the f_v data. The predicted post incubation f_v' is \approx 0.025%/dpa, which is about 25% of the measured value. However, given the sensitivity of f_v' to the sink strength ratios shown in Figure 10b, such differences are not entirely surprising. An improved GTF&C master model is under development that will be reported in the next FSA, prior to submission for journal publication.



Figure 12. a) ISHI, DII and literature $f_v(dpa - dpa_i)$ along with the common fit curve; and, b) the $f_v(dpa)$ model for He/dpa=10 appm He/dpa along with ±50% uncertainty estimates.



Figure 13. The he GTF&C void swelling model for a) ISHI; and, b) DII experiments [30].

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1.6 A STUDY ON THE STABILITY OF ALPHA-PRIME (α ') **PRECIPITATES IN HIGH PURITY Fe-18Cr ALLOYS AFTER ION IRRADIATION**—Y. Zhao, S.J. Zinkle (University of Tennessee), A. Bhattacharya (Oak Ridge National Laboratory), J. Henry (CEA)

OBJECTIVE

The objective of this work is to study the stability of α ' precipitates after heavy ion irradiation at different combinations of temperature, dose and dose rate conditions. The final size and distribution of α ' clusters are influenced by both radiation enhanced diffusion (mainly controlled by temperature, with some influence of dose rate) and ballistic dissolution (mainly controlled by dose rate and bombarding ion details). By quantifying the properties of α ' clusters over a range of irradiation temperatures and dose rates, a better understanding of phase stability in the Fe-Cr alloy system can be obtained.

SUMMARY

Ultra-high purity Fe-Cr alloys with 10-18 wt.% Cr were irradiated with 8 MeV Fe ions to a midrange (~1 um) dose of 0.35-3.5 displacements per atom (dpa) between 300-450 °C at 10⁻³, 10⁻⁴ and 10⁻⁵ dpa/s. Following irradiation, atom probe tomography (APT) was employed to characterize the number density, radius, volume fraction and Cr concentration of Cr-rich clusters. Homogeneously distributed α ' precipitates were revealed when radiation-enhanced diffusion dominates over ballistic dissolution.

PROGRESS AND STATUS

As noted in a prior report [1], high purity Fe-Cr alloys with 0-25 wt. % Cr were received from CEA, France and TEM - shaped specimens were prepared and irradiated with 8 MeV Fe ions in the Michigan ion beam laboratory (MIBL). The ion-irradiated Fe-14Cr and Fe18Cr specimen conditions summarized in Table 1 have been examined. A high density of nanoscale Cr-rich precipitates was generated during irradiation of the initial solid solution alloy for all examined irradiation conditions.

Table 1. Ion irradiation conditions for which the APT characterization was summarized in a prior report [1]

Mid-range / Peak dose (dpa)	Fluence (ions.cm ⁻²)	Flux/mid-range dose rate (ions.cm ⁻² .s ⁻¹ / dpa.s ⁻¹)	Temperatures (°C)	Estimated time (h)
0.35/ 0.84	8.83x10 ¹⁴	2.52x10 ¹¹ /10 ⁻⁴	350 450	0.97
0.35/ 0.84	8.83x10 ¹⁴	2.52x10 ¹⁰ /10 ⁻⁵	450	9.72
3.5/ 8.4	8.83x10 ¹⁵	2.52x10 ¹¹ /10 ⁻⁴	350 450	9.72

In order to evaluate the critical conditions for α' precipitate formation or dissolution, in the present study the Fe-18Cr samples were irradiated by 8 MeV Fe ions at 4 additional conditions given in Table 2 to investigate either lower temperature or higher dose rate. These irradiation conditions were selected to boost the relative importance of ballistic dissolution and/or to suppress the radiation enhanced diffusion processes. In order to identify the critical irradiation conditions where α' precipitate formation did not occur. Part of the irradiation APT characterization has been completed and the results will be summarized here.

Mid-range / Peak dose (dpa)	Fluence (ions.cm ⁻²)	Flux/mid-range dose rate (ions.cm ⁻² .s ^{.1} / dpa.s ^{.1})	Temperatures (°C)	Estimated time (h)
0.35/ 0.84	8.83x10 ¹⁴	2.52x10 ¹² /10 ⁻³	300 350 450	0.097 (5min50s)
0.35/ 0.84	8.83x10 ¹⁴	2.52x10 ¹¹ /10 ⁻⁴	300	0.97

Table 2. Summary of 4 additional ion irradiations performed on Fe-18Cr in the present study

Experimental Procedure

This report focuses on the irradiation reaction of Fe-18Cr alloy. The specimen's chemical composition, sample reparation procedures, and details related to ion irradiation experiments have been given in the prior report [1].

The precipitates in the irradiated samples were characterized by the LEAP 4000X HR model APT located Center for Nanophase Materials Sciences (CNMS) at the Oak Ridge National Laboratory (ORNL). The samples for APT analysis were prepared by focused ion beam (FIB). All the datasets were collected with voltage mode for the sake of higher spatial resolution. The Cr-enriched α ' precipitates are identified and quantified through the cluster analysis algorithm inherent to the IVAS software. For better comparison, results presented in the previous reports are also shown here.

Results

Figure 1 compares the effect of dose rates for Fe-18Cr irradiated at 450 °C to a total dose of 0.35 dpa. The as-received Fe-18Cr result is also shown to give the Cr distribution before ion irradiation. All the pink points denote the Cr atoms. The Cr atoms are distributed quite homogeneously before irradiation, so there are no pre-existing α' precipitates. The clustering of Cr atoms was observed at all 3 dose rates, even at the relatively high 10⁻³ dpa/s dose rate. Figure 2 plots the change of radius, number density, Cr concentration in precipitates and matrix and the volume fraction of α' precipitates. The radius and number density for the three dose rates are like each other and within the statistical error. The dose rate of 10⁻⁴ dpa/s leads to the highest Cr concentration in the clusters, lowest Cr concentration in the matrix, and the highest volume fraction for the clusters. By checking the precipitate size distribution, this dose rate also gives the most broadened distribution, which indicates more clusters have entered the growth stage.



Figure 1. Distribution of Cr atoms before irradiation (a) and after ion irradiation at 450 °C to 0.35 dpa at 3 different dose rates: (b) 10⁻⁵ dpa/s, (c) 10⁻⁴ dpa/s, (d) 10⁻³ dpa/s.



Figure 2. Quantitative results for α ' precipitates after ion irradiation (a) the radius of the clusters, (b) number density, (c) Cr concentration in the clusters, (d) Cr concentration in the matrix, (e) volume fraction of the precipitates.

Figure 3 presents the distribution of α' precipitates after irradiation to 0.35 dpa at 300 to 450 °C and with dose rates of 10⁻⁴ dpa/s. For better visibility, the background is filled with blue points that correspond to Fe atoms in order to provide the shape of the APT tips. All the Cr- enriched clusters are indexed with different colors in order to improve visibility of adjacent clusters. A high density of nanoscale Cr-rich precipitates are visible for all examined temperatures. As summarized in Figure 4, the radius and number density of the clusters are very close (within the statistical error) for all 3 temperatures. Cr concentration in the precipitates and the matrix monotonically increases and decreases, respectively as temperature increases. The volume fraction of α' phase exhibits the same trend as temperature. The size distribution of clusters indicates that the clusters formed at 300 or 350 °C are still in the nucleation stage, while the precipitates form at 450 °C has entered the growth region. This trend is consistent with the faster diffusion process at higher temperatures (enhanced renucleation and coarsening.)



Figure 3. Distribution of Cr-enriched clusters after ion irradiated at 3 different temperatures: (a) 300 °C, (b) 350 °C, (c) 450 °C.



Figure 4. Numerical results for Fe18Cr irradiated at 3 different temperatures.

The effect of dose was investigated for Fe18Cr irradiated at both 350 °C and 450 °C at a fixed dose rate of 10^{-4} dpa/s. The size distribution of the clusters at both temperatures is given in Figure 5. A slight shift of cluster size distribution to larger size is observed at 350 °C, and there is only one peak in this distribution, which indicates that all the clusters are still in the nucleation stage. For the clusters formed at 450 °C, they are distributed more broadly for both doses compared to the 350 °C size distribution, and the broadening of more pronounced at the higher dose. This suggests that a higher fraction of clusters have entered the growth stage at 450 °C after 3.5 dpa compared to 0.35 dpa.





Conclusion

The formation of α ' precipitates was observed in Fe-18%Cr for all investigated ion irradiation conditions (300-450°C, 10-5 to 10-3 dpa/s). Overall, the radiation enhanced diffusion prevails over ballistic dissolution effects for all the investigated conditions.

3 parameters on the final cluster formation were investigated:

- Dose rate: at 450 °C, 10⁻⁴ dpa/s is most effective for Cr clustering compared to 10⁻⁵ and 10⁻³ dpa/s.
- Temperature: at 10⁻⁴ dpa/s, the α and α' phase separation kinetics increases monotonically with increasing temperature because radiation enhanced diffusion is monotonically increased.
- Dose: at 450 °C, increasing dose causes moderate growth of α' precipitates as evidenced by the broadened precipitate size distribution at the higher dose. At 350 °C, the precipitates appear to be still in the nucleation stage up to 3.5 dpa since no significant broadening is observed.

Future Work

- The APT characterization will be performed on thermally aged Fe-18Cr (100 to 900 h at 475 °C) and pristine Fe-18Cr and Fe-14Cr samples before and after irradiation at higher dose rates and/or lower temperatures.
- The collected APT data will be further analyzed to correct the trajectory aberration and to separate the connected clusters to get a more precise value for the Cr concentration in clusters. The effect of C, N or dislocations on the precipitate number density and volume fraction will be evaluated.
- The effect of Cr concentration on the α ' precipitates population will be studied.

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2. ODS AND NANOCOMPOSITED ALLOY DEVELOPMENT

2.1 NOVEL INSIGHTS INTO NEUTRON IRRADIATED ODS ALLOYS BY ADVANCED CHARACTERIZATION—A. Bhattacharya, A.R. Lupini, D.T. Hoelzer, J.W. Geringer, Y. Katoh (Oak Ridge National Laboratory), S.M. Levine, S.J. Zinkle (University of Tennessee)

OBJECTIVE

To understand the irradiation tolerance of oxide dispersion strengthened (ODS) alloys for fusion environments, high dose (>60 dpa) neutron irradiation at 300 °C was performed on three alloys, 20%Cr base PM2000, 14%Cr base MA957 and 12%Cr base 12YWT, at HFIR. These irradiations were a part of the JP28-29 campaign, with irradiations lasting over eight years. The objective of this project is to use analytical and high-resolution scanning transmission electron microscopy (HRSTEM) to characterize the morphology, crystallography, and chemistry of nano-dispersoids in the ODS alloys.

SUMMARY

Advanced analytical electron microscopy was performed using LAMDA and CNMS facilities on three ODS alloys irradiated to 60-80 dpa at ~300 °C, with focus on the PM2000 alloy. Aberration-corrected HRSTEM, high-resolution electron energy loss spectroscopy (HRSTEM-EELS), as well as high throughput energy dispersive X-ray (EDX) spectroscopy revealed dramatic evolution of the oxide particles caused by the neutron irradiation. The initially crystalline oxide particles transformed into "sponges". These sponges consisted of crystalline islands of iron and chromium embedded in amorphous yttrium-aluminum-oxygen rich shells. Numerous particles developed internal cavities.

PROGRESS AND STATUS

Nanostructured ODS alloys, which consist of a high density of oxide nanoparticles embedded within a ferritic/martensitic steel matrix, are the most promising candidate materials for the first wall and blanket structure of fusion reactors. Inside fusion reactors, ODS alloys will suffer unprecedented radiation damage by 14 MeV neutrons (~150-200 dpa) over a wide range of irradiation temperatures (~300 - 700 °C) [1, 2]. It is because the oxide particles in ODS are crucial to the alloy's overall performance that their stability under neutron irradiation is of key importance. Evidence for the good radiation tolerance of oxide particles in ODS alloys up to high doses by ion irradiations has emerged recently [3]. However, despite the scientific interest, ODS alloys irradiated to high neutron doses have not been available for in depth analysis until recently. As a result, little is known about the radiation tolerance of oxide phases when irradiated using neutrons. To address this key knowledge gap, various ODS alloys (PM2000, MA957 and 12YWT) were irradiated in HFIR as a part of the JP28-29 irradiation campaign to doses greater than 60-80 dpa at ~300 °C. The material for this study is PM2000, a high Cr alloy where the native dispersoids are YAG/YAM phases [4].



Figure 1. HAADF imaging and STEM-EDX maps of oxide particles in unirradiated PM2000 alloy.

In the unirradiated state, high-angular annular dark field (HAADF) imaging and EDX mapping in FEI Talos STEM revealed the chemistry of the particles which were primarily Y, AI and O rich. Interestingly, all the particles exhibited a Ti shell structure (Figure 1). Aberration corrected STEM imaging using Nion Ultra-STEM 200 at CNMS revealed that all the particles were fully crystalline in the unirradiated state, as can be seen in Figure 2 where the atomic columns inside an oxide particle are imaged. Typical diameter of particles was > 30 nm.



Figure 2. Aberration corrected STEM-HAADF imaging of an oxide particle in unirradiated PM2000 alloy revealing full crystallinity.

After neutron irradiation to >60 dpa at ~300 °C, a combination of EDX mapping, aberration corrected highresolution EELS and HAADF imaging revealed that the Y-AI-O particles experienced extensive degradation. The neutron irradiation transformed the initially crystalline oxide particles into "sponges". These sponges consisted of islands of Fe/Cr rich regions embedded in Y-AI-O rich shells. Numerous particles also developed internal cavities. Examples of degraded particles with internal cavities and Fe/Cr islands are shown in Figure 3 HAADF image and EDX maps.



Figure 3. The HAADF imaging and STEM-EDX maps of oxide particles in neutron irradiated PM2000 alloy revealing the evolution of the particles into sponge-like structure.

To better understand the structure of the Fe/Cr islands and Y-Al-O rich shells, HRSTEM was performed on the irradiated alloy. The corresponding HAADF images are given in Figure 4 where the atomic-scale structure of the sponge-like particles was imaged. These experiments revealed that the Fe/Cr islands were fully crystalline, while the Y-Al-O shell was amorphous. The Fe/Cr rich islands were bcc in structure and had a different crystallographic orientation as compared to the Fe-Cr-Al rich matrix. This can be clearly seen in the high magnification image of the island in Figure 4 where the different regions are annotated.

This behavior suggests that nanoscale decomposition of the initially single-phase oxide particles has occurred, followed by amorphization of a portion of the two-phase particles. Further, no Ti shell existed after the irradiation. Such complex and dramatic evolution of the oxide phases under irradiation was unexpected and necessitates an improvement in fundamental understanding of dispersoid stability under irradiation.



Figure 4. Atomic-scale HAADF imaging of oxide particles in neutron irradiated PM2000 alloy revealing the structure of the particles, crystallinity of the Fe/Cr rich islands and the amorphous Y-AI-O shell.

Next steps are to characterize these nano-oxides using 3D atomic electron tomography and EDX tomography to better understand the structure of these particles. These experiments will be performed as an accepted user proposal in the Molecular Foundry [5] lab of Lawrence Berkley National Lab (LBNL). Further, to better understand the evolution of the crystalline oxides into such complex sponges, low dose (~5-20 dpa) HFIR irradiated PM2000 alloy and other ODS alloys will be characterized using similar characterization techniques as in the present study.

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2.2 RESISTANCE TO VOID FORMATION IN ODS ALLOYS IN THE PRESENCE OF IN-SITU HELIUM INJECTION—Danny J. Edwards, Karen Kruska, Jing Wang, Charles Henager, Richard Kurtz, Wahyu Setyawan (Pacific Northwest National Laboratory), T. Yamamoto, Y. Wu and G.R. Odette (University of California-Santa Barbara)

This is an Extended Abstract for a Presentation presented at the 19th International Conference on Fusion Reactor Materials

The ODS steels are a candidate structural material in fusion reactors due to their great ability to withstand the high temperatures and neutron irradiation environment. It is hypothesized that finely dispersed oxide clusters act as sinks for helium formed in the structural components of a fusion reactor. The large number of densely distributed clusters hinders the formation of any large helium bubbles by trapping the helium into smaller bubbles at a higher number density. In order to study this, a series of irradiation experiments [1,2] aimed at documenting the effects of in-situ helium injection on a variety of 16 different ferritic/martensitic alloys, oxide dispersion strengthened (ODS) alloys and nanostructured ferritic alloys (NFAs) was conducted in the High Flux Isotope Reactor (HFIR). The helium was injected via the ⁵⁹Ni transmutation of a NiAI coating on TEM discs of each alloy, which produces a 4.7 MeV alpha particle that is injected into the adjacent alloy over a uniform depth up to 6-8 µm while simultaneously being irradiated with neutrons at 3.9, 9 and 21 dpa from at fusion relevant temperatures (300, 400 and 500°C). Post-irradiation examination has been conducted jointly at UCSB and PNNL and is still ongoing as we work our way through the various irradiation conditions and materials. The helium injection rates varied with coating thickness and location in the subassembly, ranging from 10, 20, 38 and 60 appm He/dpa. In this presentation we discussed how the oxide particle distribution affected the formation of voids and helium bubbles in the ODS alloys 14YW and PM2000, and the NFA 14YWT and 12YWT. In some alloys, direct association between cavities and dislocations was observed, so some discussion of the relationship between these two features was presented and discussed. Characterization was performed using analytical TEM/STEM analysis on FIB milled lamella taken from both the helium-injected side of the TEM foil, and from the backside of the foil that nominally experienced only neutron irradiation with comparatively low levels of helium (1 appm/dpa). The type, size and spatial distribution of the oxide particles proved to be critical in determining the resistance to void formation. This is best illustrated in Figure 1, showing the comparative cavity sizes and densities measured for 3 different alloys PM2000, 14YW, and 14YWT.



Figure 1. Cavity size distributions are provided for PM2000, 14YW and 14YWT irradiated with neutrons to 21 dpa at 500°C with a total helium of 1230 appm. 14YW exhibited the worst swelling resistance, producing a high density of voids up to 10 nm in diameter that formed on particle interfaces. The PM2000 and 14YWT formed primarily bubbles associated with either large dislocation loops or nano-oxide particles, respectively. In the PM2000, the only voids were located inside the low density of oxide particles.

The analysis showed clearly that the oxide particles and/or their interfaces serve as effective trapping sites for the helium, allowing small bubbles to form. If the oxide distribution is too coarse, as in the case of the 14YW alloy, then each bubble is able to capture enough helium to transition into an unstably growing void, which occurs when the bubbles reach ~3 nm in diameter. The 12YWT and 14YWT, which contains small additions of Ti to refine the particles into a Y₂Ti₂O₇ phase <3 nm in diameter at a density greater than 10²³ m⁻³, exhibited no voids beyond those associated with a low density of TiN particles and much larger intergranular oxycarbonitrides induced during processing. PM2000 stood out from the other alloys because the particle distribution (~10²¹ m⁻³, 30 nm avg diameter) allowed void formation inside each particle, but within the matrix, a fine distribution of helium bubbles formed on the high density of arrayed dislocation loops, yielding a bubble distribution similar to the NFA 14YWT and 12YWT.

The results of this experiment demonstrated the effectiveness of a high density ($\sim 10^{23}$ m⁻³) of stable oxide particles with diameters of <3 nm in maintaining the swelling resistance of ferritic alloys up to high doses and high helium levels when irradiated at 500°C. Experimental work will focus on the same alloys irradiated at 400°C to comparable doses and helium levels, helping to fill in a gap in the radiation effects database on alloys of these types.

Acknowledgements

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2.3 INFLUENCE OF CLUSTER ANALYSIS PARAMETERS ON PERCEIVED SIZE OF ODS PARTICLES IN NEUTRON IRRADIATED AND He IMPLANTED 14YWT—Karen Kruska, Jing Wang, Danny J Edwards, Richard J Kurtz, Wahyu Setyawan (Pacific Northwest National Laboratory)

OBJECTIVE

The objective of this research is to employ atom probe tomography (APT) to study the effect of neutron irradiation and helium on the composition and distribution of oxide particles and formation of precipitates in oxide dispersion strengthened steels and nanostructured ferritic alloys.

SUMMARY

In the previous report we presented APT data that suggested that ODS particles in the as-received, neutron irradiated, and He-implanted samples of 14YWT all had a mean particle size (diameter) of 4-5 nm and in each case a significant number of particles were larger than 5 nm. Particles of this size range would be expected to be readily characterized with TEM EDS. In our own TEM analysis we found that we could not readily detect the ODS particles using STEM EDS. This suggests that a second look at the cluster analysis performed on the APT data is necessary. To calculate the precipitate sizes in the atom probe data in the previous report, the positions of cluster ions – Y, TiO and CrO – in the data set were assigned to a cluster and the cluster size was calculated with the assumption that the clusters are spherical. In this report, we test the stability of the clustering algorithm calculating cluster size after cluster search with three sets of different clustering ions. Furthermore, the cluster diameter is calculated in three different ways. The first two start with the position of cluster ions and assumes a close packed stoichiometric structure (Dp). While D(sphere) and Dg are insensitive of different (sensible) selections of cluster ions, Dp is expected to be sensitive to ion selection. Our initial analysis shows that Dp is likely smaller than the real cluster size but is a sensible lower limit of the predicted particle size.

PROGRESS AND STATUS

The ODS steels are a candidate structural material in fusion reactors due to their great ability to withstand high temperatures. It is hypothesized that finely dispersed oxide clusters act as sinks for helium formed everywhere during the operation of a Tokamak style fusion reactor. The large number of densely distributed oxide clusters hinders the formation of any large helium bubbles and subsequent embrittlement. The desired ODS particle size in a suitable alloy is 2-5 nm. Such small particles dispersed throughout a 20-100 nm thick matrix are difficult to see with TEM based microscopy and spectroscopy techniques and even more difficult to accurately measure and count [1]. Atom-probe tomography is much more suited to detect these particles, but the data reconstruction comes with its own challenges. While the ion count in each particle comes directly from the measurement, particle size and shape in the visualization may vary depending on the reconstruction parameters chosen. In this report, we performed APT study of 14YWT samples. In collaboration with UCSB the samples were in-situ helium injected (ISHI) and irradiated in HFIR at 500 °C up to 21 dpa where one side of the samples was neutron-irradiated only while the other side also received He injection up to ~1230 appm He.

Hypothesis: ODS particles in 14YWT in all three conditions are < 5 nm in size.

Experimental Procedure

A novel cluster search algorithm developed at PNNL (OPTICS) [2] was used to quantitatively evaluate ODS particles. Cluster searches were conducted using three sets of ions:

1. Ti-Y-O set (Ti, TiO, TiO2, Y, YO, O).

- 2. Simple set (TiO, Y, YO, CrO). These are the most common ions in clusters.¹
- 3. Cluster ion set (Ti, TiO, TiO2, TiN, Y, YO, Si, SiO, SiO2, CrO, CrN, FeO, O, N). These are all the ions that are a lot more common within a 2% Y isosurface than in the matrix.²

The diameter was calculated assuming that particles are spherical (D(sphere)) and from the radius of gyration (Dg) for each case. A third way to calculate the cluster size was explored. As the traditional techniques to calculate size are affected by trajectory aberrations occurring during APT analysis, we calculated cluster size based on corrected ion count only. All ions containing Y, Ti and O were counted in each cluster. The ions were decomposed into atoms and all atoms were counted. As the detector efficiency is ~37%, atom count was corrected for detector efficiency. The total mass of each cluster was calculated assuming each cluster is stoichiometric $Y_2Ti_2O_7$, i.e. each 11 atoms have a combined mass of 385.539 Da or 6.404 x 10 ⁻²² g. $Y_2Ti_2O_7$ has a density of 4.95 g/cm³ or 4.95 x 10 ⁻²¹ g/nm³ [3]. With this information, the volume, radius and diameter of each (spherical) cluster were determined.



Figure 1. Diameter of gyration vs diameter of spherical cluster in A) as-received, B) neutron-irradiated, and C) He-injected samples of 14YWT. D) Comparison of traditional diameters with calculated close packed cluster diameter.

 $^{^{1}}$ Cr is common as well, but there is too much Cr in the matrix and in the irradiated sample it would be difficult to distinguish α' and ODS.

² Cr was excluded here as well.

Results

Figure 1 shows the results of the comparison in clustering parameters. It is immediately obvious that the choice of cluster ions did not have a big influence on resulting cluster size with either of the traditional methods to calculate cluster size. This is because additional ions chosen are likely located between the most common ions and hence do not influence the outer "cluster boundary".

Both the diameter of gyration and the diameter of an assumed spherical cluster are larger than what is expected from TEM analysis (~2 nm) [4]. To overcome the challenge posed by trajectory aberrations in APT analysis an additional diameter was calculated based on ion count and cluster density alone (Figure 1 D). As the density of stoichiometric $Y_2Ti_2O_7$ was used for the calculation, only the clustering data for the Ti-Y-O cluster ion set was deemed suitable. The results show that the clustering diameter in this case is about 50% smaller and closer to what is expected.

The close-packed diameter can be considered a minimum cluster size as all ions considered here were detected in the APT dataset. Compositional analysis of some larger clusters has shown Si and N associated with ODS clusters and it is likely that some amount of matrix components is present also. As the resulting close-packed diameter is directly proportional to the number of atoms assigned to the cluster rather than their position in the reconstructed dataset, this method of size determination is expected to be much more dependent on the set of ions selected for the cluster search.

Future Research

The close-packed diameter method predicts particle size < 2 nm, which seems to be consistent with the difficultly of these particles to be observed under TEM, indicating that the close-packed diameter offers a reasonable method for calculating cluster sizes. However, more analysis needs to be conducted to ensure the correct oxide phase was selected for density selection. Partial occupation of the oxygen sublattice in a certain phase (such as in $Y_2Ti_2O_7$) may also affect the calculated diameter, nevertheless this effect is expected to be small given that O is significantly lighter than Y or Ti. Furthermore, inclusion of additional "impurity" cluster ions needs to be further explored and possibly adjusted for each irradiation condition.

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2.4 APPLICATION OF FLASH POLISHING AND DCI-STEM DISLOCATION IMAGING ON CHARACTERIZATION OF DEFECTS—Dalong Zhang, Danny J. Edwards, Alan Schemer-Kohrn, Charles H. Henager Jr., Wahyu Setyawan (Pacific Northwest National Laboratory) T. Yamamoto, Y. Wu and G.R. Odette (University of California-Santa Barbara)

OBJECTIVE

The objective of this work is to apply the flash polishing technique to remove artifacts and damages caused by focused ion beam (FIB) preparation of transmission electron microscopy (TEM) specimens for neutronirradiated materials. Subsequently, diffraction contrast imaging in scanning transmission electron microscopy (DCI-STEM) is under development to clearly image dislocation lines and loops in flash polished specimens. The combined application of flash polishing and STEM dislocation imaging will be extended to study in-situ helium injected materials, then compare with neutron-irradiated ones in defect types, densities, and possible evolution of particles and precipitates.

SUMMARY

Neutron-irradiated PM2000 alloy was milled and thinned by standard FIB procedure for TEM specimen preparation. The TEM examination of as-FIB specimens revealed typical FIB damage of black spots and Moiré fringes. In-house developed flash polishing technique was then used to remove damage in as-FIB specimens. Preliminary TEM/STEM results showed that without FIB damage and artifacts, line dislocations and loops gave rise to much clearer contrast, enabling better defect analysis and quantification. In addition, comparison was made between conventional TEM under two-beam condition and DCI-STEM using a FP specimen. While the former was able to image dislocations and loops with diffused contrast, DCI-STEM provided much sharper contrast under both bright field and dark field conditions.

PROGRESS AND STATUS

Introduction

In addition to neutron-irradiation induced or enhanced defects, including dislocation loops and precipitates, metallic materials used in a fusion reactor are also faced with high concentration of helium leading to helium embrittlement [1, 2]. In order to understand helium effects, in-situ helium injection (ISHI) experiments were carried out on a family of fusion relevant materials, including Eurofer97, 14YWT, MA957, PM2000, etc. [3-5]. While some materials, e.g., Eurofer 97 and 14YWT, might serve as future/near-future candidate materials for building fusion reactors, others serve as "model" materials, e.g., PM2000, to obtain fundamental understanding on helium effects. Previous work has focused on the characterization of the samples that were neutron irradiated in HFIR at 773 K (the 500C samples). Now, we start to examine the 400C samples, particular interests are on 14YWT, 14YW, and PM2000.

For TEM characterization of dislocation loops and helium bubbles, specimens are often prepared by FIB milling. However, FIB milling itself is actually an ion sputtering/implantation process, causing a surface damage layer up to 20 nm in thickness on both sides of the TEM specimen [6]. The FIB damage can obscure intrinsic nanoscale features especially in irradiated materials. Therefore, before in-depth characterization of dislocation loops, helium bubbles, and other features, it is highly desirable to remove these damages in FIB prepared specimens by means of flash polishing (FP) [6, 7]. In addition to FP method for careful specimen preparation, advanced STEM based techniques have been developed or promoted in recent years for imaging dislocation lines and loop, helium bubbles, nanoscale precipitates, etc. [8-10]. The STEM based imaging has the advantages of bypassing thickness contrast/bending contours [10], higher resolution when using an aberration-corrected (AC) microscope, and coupling with spectroscopic techniques to reveal chemical information [8]. Our goal is to combine meticulous TEM specimen preparation method of FIB and FP, together with STEM based defect imaging, to better understand the relationship between neutron-irradiation induced defects and helium effects.

Experimental Procedure

Ferritic PM2000 alloy (Plansee GmbH) was machined into discs of 3 mm diameter. The discs were coated with ~4 µm thickness of NiAl on one side, then irradiated in High Flux Isotope Reactor (HFIR) up to ~21 dpa at 573K, 673K, 773K, respectively. Up to ~1230 appm of helium was injected on the NiAl coated side (i.e., ISHI side), whereas the uncoated side only experienced neutron irradiation ("neutron irradiated only", i.e., NIO side). Details regarding the ISHI technique have been reported elsewhere [3-5].

In this report, for exploring the application of flash polishing and DCI-STEM technique on defect characterization, we used the PM2000 samples. The PM2000 TEM specimens for both the ISHI side and the NIO side were first prepared with a standard FIB procedure using an FEI (now Thermo-Fisher Scientific) Quanta 3D FIB. The TEM examination of these as-FIB specimens, as well as flash polished specimens, was done on a cold field-emission JEOL ARM200CF microscope operated at 200 kV, equipped with a hexapole type probe Cs-corrector (CESCOR, CEOS). Figure 1 shows the in-house setup for flash polishing, with a special in-house built relay timer and a 30 V power supply on the left, a Pyrex beaker in an ethanol and dry ice cooling bath in the middle, and an Au-coated tweezer holding an Au grid where a FIB lift-out specimen is mounted to. Further details regarding the in-house flash polishing setup has been reported by Schemer-Kohrn et al. [11]. To date, a TEM specimen for the NIO side was successfully flash polished and examined by TEM and STEM, whereas work is ongoing to flash polish and examine TEM specimen for the ISHI side. To facilitate meaningful comparison, results and discussion will be focused on NIO side specimens in as-FIB condition and flash polished condition, respectively.



Figure 1. Beaker polishing setup for flash polishing FIB specimens mounted on an Au coated grid. The grid is held by the tweezers placed inside a plastic cap; the cap is inserted over the beaker [11].

Results

Figure 2 shows typical STEM micrographs for as-FIB specimen from NIO side of PM2000. Particularly, Figure 2(a) is DCI-STEM mode, analogous to g=011 two-beam condition in TEM. Black-white undulations of contrast, i.e., Moiré fringes, are present. Figure 2(b) is STEM imaging at [001] zone axis. In both cases, extra "black spots" are visible which are likely FIB damages. It is noted that DCI-STEM often gives rise to sharper dislocation contrast as compared to the diffuse contrast in the case of on-zone STEM [10]. Conversely, on-zone STEM does have the advantage of capturing both a/2(111) type and a(100) type loops without any statistical biasing from the extinction criteria [8]. Further parametric investigation on imaging conditions, including camera length, convergence angle, collection angle, etc. is ongoing to decide either DCI-STEM or on-zone STEM is better suited for defect imaging especially for flash polished specimens.

After flash polishing the specimen in Figure 2, g=011 two-beam condition CTEM imaging was first used to examine the flash polishing quality (shown in Figure 3). Looking at areas that are free of any dislocations or loops, one can see that flash polishing indeed removed most of the Moiré fringes and extra black spots. However, CTEM still has the issue of diffused dislocation contrast, which necessitated DCI-STEM.



Figure 2. Typical micrographs of PM2000 NIO side before flash polishing. (a) The DCI-STEM mode, analogous to g=011 two-beam condition in TEM. Black-white undulations of contrast, i.e., Moiré fringes, are present. (b) The STEM imaging at [001] zone axis. Extra "black spots" are visible which are likely FIB damages.



Figure 3. The g=011 two beam condition CTEM imaging for PM2000 NIO side after flash polishing. (a) Lower magnification; (b) Higher magnification.

Figure 4 represents typical DCI-STEM micrographs of PM2000 NIO side after flash polishing. Figure 4(a) is bright field, whereas Figure 4(b) is dark field obtained with an angular dark field detector. It is clear that DCI-STEM provided a much sharper dislocation contrast as compared to CTEM imaging previously shown in Figure 3. Besides dislocation line and loops, precipitates can readily be seen in STEM, like the one at the bottom-right corner. In addition, the nano-sized distinct "black dots" might also be intrinsic features in PM2000 after neutron irradiation, in the form of embryonic dislocation loops or nano-precipitates/clusters. Further high-resolution DCI-STEM and STEM-EDS study is ongoing.



Figure 4. Typical DCI-STEM micrographs of PM2000 NIO side after flash polishing. (a) bright field, (b) dark field using the angular dark field detector.

Future work includes specimen preparation and defect characterization for the PM2000 ISHI side as well as extending the characterization to the 14YWT and 14YW. Note that besides common dislocation loops and possibly precipitates, the ISHI side would also contain helium bubbles. Over focus-under focus technique in CTEM has been the standard practice for imaging helium bubbles. However, based on reciprocity theorem between STEM and TEM [8], it is postulated that STEM can also be used with the same over focus-under focus routine.

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3. CERAMIC COMPOSITE STRUCTURAL MATERIAL DEVELOPMENT

3.1 PROGRESS IN DEVELOPMENT OF SIC-BASED JOINTS RESISTANT TO NEUTRON IRRADIATION—Takaaki Koyanagi, Yutai Katoh (Oak Ridge National Laboratory), Tatsuya Hinoki (Kyoto University), Charles Henager (Pacific Northwest National Laboratory), Monica Ferraris (Politecnico di Torino), Salvatore Grasso (Southwest Jiaotong University)

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This study fills a knowledge gap regarding the neutron-irradiation resistance of SiC joints for fusion reactor applications by investigating high-dose neutron irradiation effects on the strength of selected joints and low-dose neutron irradiation effects on recently developed joints fabricated by state-of-the-art methods. An important property demonstrated was the retention of strength in SiC joints fabricated by pressure-assisted liquid-phase sintering of SiC, Ti-Si-C powders reaction sintered with hot-pressing, and calcia-alumina glass ceramics bonded by pressure-less joining after all were subjected to neutron irradiation at ~500°C to 20×10^{25} n/m² (E >0.1 MeV) for ~350 days in the High Flux Isotope Reactor. This demonstration encourages the further development of SiC for fusion applications. In addition, this study found that low-pressure liquid-phase-sintered SiC and pressure-less reaction-sintered Ti-Si-C powders also retained their strength following lower-dose irradiation at 500 and 700°C to $2-3 \times 10^{25}$ n/m² (E >0.1 MeV). Such pressure-less methods will be useful for fabricating large and/or complex-shape components.

HIGH HEAT FLUX MATERIALS AND COMPONENT TESTING 4.

4.1 FRACTURE TOUGHNESS OF ROLLED 90W7Ni3Fe TUNGSTEN HEAVY METAL

ALLOY—M.E. Alam, G.R. Odette (University of California Santa Barbara), C.H. Henager, W. Setyawan (Pacific Northwest National Laboratory)

OBJECTIVE

The objective of this research is to understand the hot-rolling effect on the room temperature fracture toughness of 90W7Ni3Fe tungsten heavy alloy.

SUMMARY

The tensile strength and precracked fracture toughness properties of the as-received four ductile phase toughened (DPT) commercially available tungsten (W)-based heavy metal alloy (WHA) composites, reinforced with 3 to 10 (wt.%) of a ductile WNiFe phase were previously characterized from room to liquid nitrogen (LN₂) temperatures. Here we explore the effect of hot rolling on the room temperature precracked fracture toughness of a similar 90WNiFe PNNL WHA, at different hot rolling reduction percentages (i.e. 0, 62, 74 and 87%). High elastic-plastic K_{Jm} toughness is observed in the 0 and 87% hot rolled alloys along with stable crack growth, while the 62 and 74% hot rolled WHA experience elastic fracture with much lower K_{Ic}. An exactly opposite trend is observed in the microhardness of the plates, which is lowest in the 0 and 87% hot rolling and highest in the 62 and 74% hot rolled condition. Possible factors confounding interpretation of these results are noted.

PROGRESS AND STATUS

Introduction

Due to high melting temperature, good conductivity, low sputtering rate and high-temperature strength, tungsten (W), and its alloys, are currently considered the most promising candidates for plasma facing component for future fusion reactor divertor applications [1–3]. This application requires that structural W-based alloys and structures that have sufficient fracture toughness to withstand the severe thermal-mechanical environment of a divertor. It is likely that monolithic W is intrinsically too brittle for this task. Tungsten heavy metal alloys (WHAs) are well known for their good room to high temperature tensile strength and ductility [4–7]. Various plastic deformation processing routes [8–16] have been used to further enhance their mechanical properties, mainly for the applications in ordnance such as kinetic energy penetrators, as well as counterbalances, etc.

Recently, WHAs have been considered as a potential structural plasma facing material for fusion reactor divertor applications [10,17-19]. To this end the precracked fracture toughness a series of as-received WNiFe (90, 92.5, 95 and 97 wt.% W with 7:3 = Ni:Fe) WHA were shown to have much higher room temperature toughness (> 10x) and much lower BDTT temperatures (-150 to -25 °C) than monolithic W (K_{IC} ≈ 8 ± 4 MPa \sqrt{m} , BDTT ≈ several hundred °C), depending on their ductile phase NiFe content [20]. PNNL has explored similar WHAs in various hot rolled conditions to characterize the effects of the composite architecture on deep notch bend bar toughness [10]. In this study, room temperature precracked fracture toughness properties of the hot-rolled 90W7Ni3Fe (wt.%) PNNL alloy at rolling reductions of 0, 62, 74 and 87% was characterized based on ASTM Standard Practice test methods [21]. The 62 and 74% hot rolled samples fractured unstably with much lower toughness of $K_{lc} \approx 35 \pm 2$ MPa \sqrt{m} (notably, still 4x higher than the monolithic W \approx 8 ± 4 MPa \sqrt{m}). The 0% and 87% hot rolled elastic plastic toughness was much higher at $K_{Jm} \approx 97 \pm 18$ MPa \sqrt{m} and $K_{Jm} \approx 118 \pm 20$ MPa \sqrt{m} , respectively, which was accompanied by extensive stable crack growth. The same toughness trends were observed in PNNLs deep notch bend bar tests. Dilatational process zone microcrack shielding was observed for the 0 and 87% hot rolled alloys, but not in the 62 and 74% deformed conditions. Microhardness shows an exactly opposite trend, with much higher hardness for the 62 and 74% hot rolled alloys ($H_v \approx 400$ kg_f/mm²), compared to the 0 and 87% WHA (Hv \approx 315 kg_f/mm²). The reason for the H_v differences is not known. While H_v difference may be a partial reason for the toughness trends, both microstructural variations (composite architectures) and the absence of differences confound easy interpretations.

Experimental Procedure

Commercially available 90W7Ni3Fe (hereafter called 90W) WHA was acquired from Mi-Tech Metals, Indianapolis, IN, USA as an as received liquid pahse sintered plate. The as-received 90W has nearly equiaxed, \approx 100% pure W particles of \approx 17 ± 7 µm size, imbedded in the continuous, honeycomb web ductile phase (DP) structure, with a composition of \approx 50%Ni-30%W-20%Fe, in units of wt.% [20]. In an effort to explore a more lamellar-like microstructures, the 90W alloy was hot rolled, to three different reductions, to produce a quasi-2D microstructure. The 90W plates were hot-rolled , in two series of hot-roll/annealing sequences. The first sequence was performed at 1,150 °C, with two passes of 5%. The alloys were then rolled at 900 °C in 11% passes to 62% and 74% reductions followed by annealing at 900 °C for 1h in a Ar-H₂ (50:50) atmosphere. Bend bar specimens from the 62 and 74% hot rolled plates were not H-degassed. The second rolling sequence was similar, started on a thicker plate that was rolled to an 87% reduction. Specimens from this plate were hydrogen de-gassed at 900°C in vacuum. Further descriptions of the processing PNNL WHA can be found elsewhere [10]. Hereafter, we will refer to the hot rolling reductions as 0R, 62R, 74R and 87R.

Figure 1a shows a schematic of rolling operation, showing the rolling related directions. The corresponding three-dimensional (3D) mesostructure of the deformed plate is also schematically illustrated in Figure 1b. Deep notch end specimens were fabricated by electrical discharge machining (EDM) at PNNL. They were ground with 220 to 2000 grit SiC sand paper to remove EDM damage and residual surface stresses at UCSB. Some specimens were then polished with 0.25µm-colloidal silica and etched in a 30% hydrogen peroxide solution for 10 min to facilitate microstructural characterization. Both optical microscopy, scanning electron microscopy (SEM) including energy dispersive x-ray spectroscopy (EDS) and electron backscatter diffraction (EBSD), were used to characterize the W particles and the surrounding NiWFe ductile phase. Details microstructural characterization procedures can be found elsewhere [20].

Room temperature Vickers microhardness measurements (H_v) were performed on the bend bar polished side surface at varying loads of 100, 200, 500 and $1000g_f$ with a 10 seconds dwell, using a LECO M-400A semi-automated hardness tester. Different loads were used to explore microhardness at various length scales relative to the micorstructure. Note, SEM was used to measure the indentation diagonals, rather than optical images, for more accurate readings. The reported average values and standard deviations were calculated in accordance with ASTM standard E834 [22], and 6 to 15 indentations were conducted per variable.

Room temperature fracture toughness tests were conducted on the small, fatigue pre-cracked, singleedge notch bend bar specimens with a nominal length (L) x width (W) x thickness (B) dimensions of 25.4 x 4.0 x 1.75 mm, oriented in the LS directions (see Figure 1a). The specimens were precracked extending from a crack length (a)-to width (W) ratios a/W =0.45 notch to a nominal a/W $\approx 0.5 \pm 0.02$ at ΔK_1 up to a maximum of 20 MPa \sqrt{m} and a load ratio R = 0.1. The specimens were heat-tinted at 400°C for 1 min to mark the pre-crack front. A three-point bend (3PB) fixture was used on a 810 MTS servo-hydraulic universal testing machine. To facilitate the in-situ optical observation of the crack tip region, the fracture specimen sides were sanded with a sequence of 2000 grit SiC followed by 9µm, 3µm and 1µm diamond lapping paper. The fracture tests were carried out at a crosshead speed of 0.04mm/ min. The ASTM standard E1921 [21] was used to calculate the elastic (K_{Ic}) and elastic-plastic (K_{Jm}) maximum load fracture toughness (a P_m in the load-displacement (P-d) curve). Three to seven specimens were tested per rolling condition.

Results

<u>Microstructure</u>

The 3D SEM micrographs of the polished and etched 0R (as-received) 90W plate shown in Figure 1c reveal randomly oriented, roughly spheroidal W particles (particle aspect ratio, PAR: 1.2 ± 0.2) surrounded by an interconnected honeycomb web structure of the ductile NiWFe phase [20]. Consistent with our previous study, the particles compositions are close to 100% W, while the NiWFe ductile phase is approximately 50%Ni, 30%W and 20%Fe [20]. However, rolled plates show highly oriented and anisotropic W-particles and highly deformed DP phases (see Figures 1d-f, 2b-c, and Table 1). For example, while the 0R plate shows roughly similar spherical particles, with approximately equal dimensions in all three directions, the 87R W particles are irregular cigar shaped pancakes, with observed dimensions of \approx 76 x 25 µm (top view) and 81x6 µm, and 29x5 µm (side and front views, respectively). Thus, the average particle dimensions are \approx 80 x 27 x 5 µm (see Figures 1c, f and 2a-c, and Table 1).



Figure 1. (a) The schematics of the rolling operations and definition of plate views, along with 3PB bend bars at LS orientation; (b) schematic of possible deformed W-particles at each view. Figure 1c-f shows the 3D representation of the rolled-plate at each view for: (c): 0R; (d) 62R; (e) 74R; and, (f) 87R, respectively.

As expected, the W particles become more elongated with increasing rolling reduction. At 87R, they are highly deformed and connected to the neighboring particles in such a way that W appears as nearly continuous strips, or lamellar structures. The DP phase was also elongated along the rolling directions. Although, there are some local microstructural inhomogeneities, the average areal percentage of DP for 0R plate is $\approx 17 \pm 4$ %, irrespective of its plate view (Table 1). The areal DP percent reduced from $\approx 14\%$ for 62R to $\approx 12\%$ for 87R for side view, while the trend is opposite for the other two views with increasing rolling reduction, resulting the average DP% for three plate views nearly similar to the as-received plate condition, following conservation of volume (see Table 1). However, the DP alloy compositions do not

vary, which is $\approx 50\%$ Ni, 30%W, 20%Fe by wt., irrespective of rolling reductions or viewing planes (see Figure 2d-i). The top and front views of the rolled plates, as shown in Figure 3, reveals that the DP phase is infused inside the elongated W particles for 87R (see Figure 3e-g), whereas that is very minimal for the 74R (Figure 3a-c), and nearly absence for the 62R plate (not shown). However, for the same plate, per se for 87R, the amount of infused DP is observed to be higher for the front view (normal view of the rolling cross-section) followed by side view and least in the top plane view. This is probably due to the tensile force that acts normal to the front view. The DP web thickness also appears thinner with the increasing rolling directions.



Figure 2. Binary black and white SEM images for the: (a) 0R at all views; (b) 87R, top; and (c) 87R, side views, respectively; representing the morphology of W particles (black) and DP phase (white). Figure 2d-e are the SE and BSE SEM images, respectively, of 87R samples used for EDS area map (f-h) and point (i) scans.

High magnification BSE SEM images also reveal orientation contrast in the deformed W particles (see Figure 3a,c,e,g). The 74R plate shows nearly micron size grains inside the particles (Figure 3c,d), whereas submicron size grains were observed for the 87R plates (Figure 3g,h). The formation of micron to ultra-fine grains might be due to the dynamic recrystallizations that occurred during hot rolling operations. Dynamic recrystallization was also observed for the DP phases with the grain size much larger (~ 4 μ m) than the W-grains inside particles (white arrows in Figure 3c). Similar observations have been reported by many researchers for similar type of WHA's [8,9,11,13]. Rolled plates, especially 87R, also shows the small W-particles fused into the DP phases (red circles in Figure 3a,e). The EBSD scans on the side surface of the deformed plate shows random texture formation [10]. However, the intensity of the texture of each plate is not known. A much finer scanning step along with a plot of misorientation angles for each plate will help to identify the nature of the subgrain structure of the W-particles and the DP phases, and their roles in mechanical properties.

90W7Ni3Fe Rolled, (%)	Views	Long axis, I, (µm)	Short axis, s, (µm)	Average, (I+s)/2, (μm)	PAR, I/s; orientation	DP (%)
0	Top, side, front	18 ± 6	15 ± 6	17 ± 7	1.2 ± 0.2; X	16.7 ± 3.6
	top	46 ± 18	26 ± 10	36 ± 13	1.9 ± 0.5; X	17.8 ± 1.1
62	side	41 ± 16	9 ± 4	25 ± 10	4.5 ± 1.8; //	14.1 ± 2.6
	front	27 ± 12	10 ± 3	18 ± 7	2.7 ± 1.1; //	18.8 ± 1.4
	top	58 ± 23	23 ± 9	41 ± 15	2.6 ± 1.0; //	19.9 ± 1.1
74	side	52 ± 23	7 ± 3	27 ± 12	7.4 ± 3.9; //	13.2 ± 1.8
	front	27 ± 11	7 ± 2	17 ± 6	3.9 ± 1.9; //	20.4 ± 1.2
	top	76 ± 31	25 ± 8	50 ± 18	3 ± 1; //	21.1 ± 1.3
87	side	81 ± 37	6 ± 2	44 ± 19	15 ± 9; //	11.7 ± 0.9
	front	29 ± 8	5 ± 2	16 ± 4	7 ± 3; //	22.6 ± 0.8

Table 1. Grain morphology and ductile phase area percent of the rolled 90WNiFe

DP = ductile phase; PAR = particle aspect ratio; X = random; // = parallel.

Microhardness

Vickers' microhardness (Hv) results for the as received and rolled 90W alloy tested at RT are shown in Table 2 and Figure 4. The average hardness increases with decreasing indentation load, irrespective of rolling reduction. At 100gf load, the indentation diagonal is $\approx 22 \,\mu$ m (see Figure 4b), which is very close, if not smaller, to the W-particles size. Thus there is a high chance that the indentation load mostly probes the W-particles, which are much harder than the DP phase, therefore, resulting in higher hardness values. At higher loads, say for 1000gf, the indentation is much deeper with a diagonal length of $\approx 76 \,\mu$ m (see Figure 4c,d), that involves both the W particles and DP phase, which therefore lowers the global hardness of the material. Except for 87R, the microhardness value also increases with increasing rolling reductions (i.e. $312 \pm 4 \, \text{kgr/mm}^2$ for 0R and 400 $\pm 14 \, \text{kgf/mm}^2$ for 74R at 500gr, see Table 2). It is expected that the dislocation density increases with increasing rolling reduction, increasing the hardness value [9,11,14,16]. Note, per the previous statement, 87R should have shown the highest hardness. In reality, the 87R shows much lower hardness (332 $\pm 7 \, \text{kgr/mm}^2$ at 500gf) than the 62R and 74R samples, and only slightly higher hardness than the 0R plate. No indentation cracking was observed in any of the test conditions, including for 1000gr on 87R plate (see Figure 4c,d for SE and BSE images of the same indentation).



Figure 3. The BSE SEM images showing (a,e) top; and (b,f) front view of the 74R and 87R plates, respectively. Figure c,d (for 74R) and g,h (for 87R) show the higher magnification images that reveal subgrain structures, indicated by red arrows (for W) and white arrows (for DP).

Table 2. Vickers microhardness (Hv) as a function of indentation loads on rolled plates					
Loads	0R	62R	74R	87R	
100gf	359 ± 13	-	-	365 ± 13	
200gf	337 ± 8	-	-	344 ± 9	
500gf	312 ± 4	395 ± 9	400 ± 14	332 ± 7	
1000gf	308 ± 8	392 ± 5	391 ± 6	317 ± 3	





Figure 4. Plot shows the RT hardness as a function of indentation loads and rolling reduction (a); indentation impression on the 87R for (b) 100gf; (c,d) 1000 gf load. The SE (c), and BSE SEM (d) images for the same 1000gf load impression do not show any indentation cracking.

Room temperature fracture toughness

The room temperature fracture toughness tests on the various rolled plates have been conducted on the fatigue precracked bend bars using a 3PB fixture. The corresponding normalized load-displacement (Pnd_n) curves are shown in Figures 5 and 6a. Here, the load is normalized following ASTM E-1820-A15 [23] as $P_n = P/{WB(1-a/W)^2}$ which is valid up to maximum load (P_m), taken to coincide with crack extension; the displacement is normalized simply diving by the span. S. Here, W and B are the width and thickness of the specimen, respectively. Note, the P_n-d_n curves shown in Figure 5 are for comparison only, and not for calculating R-curves. As reported previously [20], all the 0R (as-received) 90W specimens show very stable crack growth with an maximum load fracture toughness, K_{Jm} ≈ 97 ± 18 MPa√m, ≈ 12 times higher than the monolithic W toughness ($K_{lc} \approx 8 \pm 4$ MPa \sqrt{m}) (see Table 3 and Figure 5a). These P-d curves have an elastic loading region, followed by a plastic yielding deviation from linearity. The subsequent increasing P is due to the growth of the plastic zone and strain hardening. The much larger decreases in P following Pm reflects stable crack growth. Note, all the tests were intentionally stopped after Pm, before final specimen fracture. The high standard deviation is due to the local microstructural inhomogeneities.

However, for the 62R and 74R specimens, only elastic loading was observed and the crack propagates very fast after reaching P_m, with a relatively low elastic fracture toughness, $K_{Ic} \approx 35 \pm 2$ MPa \sqrt{m} (see Table 3 and Figures 5b,c and 6d,e); note this is still more than 4 times higher than the pure W. In contrast the 87R specimens showed very stable crack growth having similar loading zone, with even higher elastic and plastic K_{Jm} \approx 116 \pm 20 MPa \sqrt{m} compared to the 0R condition (see Table 3 and Figures 5d, 6f).



Figure 5. Normalized room temperature load-displacement (P_n - d_n) curves for the (a) 0R; (b) 62R; (c) 74R; and, (d) 87R 90W plates, respectively. Note, all the 62R and 74R specimens are fractured unstably, whereas all 0R and 87R specimens show stable ductile tearing.

The representative normalized P_n -d_n curves for all the rolled plates are also shown in a Figure 6a, and their corresponding K_{Ic} and K_{Jm} are plotted in Figure 6b along with the average H_v values for 500 and 1000g_f. Notably the H_v of the rolled plate exactly the opposite trends compared to the K_{Ic} and K_{Jm} values (see Table 3 and Figure 6b). The P_m for the 62/74R plates are marginally higher than the other two plate conditions (see Figure 6a). Neither the hardness nor toughness trends are understood.

Our previous study [20], observed that the side surface view of all room temperature 0R specimens showed large numbers of particle-sized W cleavage (WC) blunted microcracks in the process-zone (see Figure 7a). Some W-W (WW) interface fracture and W-NiWFe interfacial debonding (WD) events were also observed. The microcracks are arrested by the NiWFe DP and blunt under increased load. Many of the cleaved and unbroken W particles also deform in the principal stress direction. All these events help to improve the toughness of the as-received 0R 90W plate. However, these events are mostly absent for the 62 and 74R specimens, as illustrated in Figure 7b-d, minimal WC near the crack propagation zone (Figure 7c) with a corresponding clean and sharp crack and microcrack free process zone (Figure 7d). In this case, once cracks initiate they link up with nearby co-planar cracks quickly, without cleaving neighboring W particles, resulting in unstable crack growth (see Figure 6a,d,e). For the 87R plate (see Figure 7e-h), all the same toughening mechanisms that occur in the 0R condition are observed. In addition, DP infused W-particles are observed for the 87R plate (see Figure 7g). One difference between the 87R and 0R plates are the microcrack orientations, where they are mostly $\approx 45^{\circ}$ and perpendicular to the principal loading directions for the 0R and 87R, respectively (see Figure 7a and f). This is due to the



random orientation of the 0R W-particles, in contrast to the highly aligned and thinned W particles in the 87R.

Figure 6. Normalized RT load-displacement (P_n - d_n) curves for the rolled plates (a); and their corresponding toughness values (b). Note, filled and half-filled squares represent stable and unstable crack propagation, respectively. Average microhardness values for the 500 and 1000gf are also plotted in Figure 6b (red diamonds). Figure 6c-f shows the side surface crack propagation for 0, 62, 74, and 87R, respectively.

Table 3. Average microhardness	(500gf+1000gf) and room ten	nperature fracture toughness

Rolled W	0R	62R	74R	87R
Hardness (kgf/mm²)	310 ± 6	394 ± 8	397 ± 12	327 ± 10
Kıc/Kյm (MPa√m)	97 ± 18	35 ± 2	35 ± 3	116 ± 20



Figure 7. Microcleavage process zone for the 0R (a), crack propagation path at different areas for 74R (b-d), 87R (e-h), respectively.

Figure 8a-d shows secondary electron SEM fractographs for the 0, 62, 74 and 87R, respectively. Almost 100% tungsten cleavage (WC) dominated local fracture modes are found for the 62 and 74R plates (Figure b,c), as observed for the low temperature elastic fracture for the undeformed particles [20]. In contrast, the 87R and 0R plates show all four well-known local fracture modes (WC, WW, DR, WD), with the dominance of W-W decohesion (see Figure 8a,d). The pre-test microstructure shows the presence of DP inside the W-particles for the 87R condition (Figure 3f), whereas they are merely seen for the 74R plate (Figure 3b). BSC SEM fractographs for the 74R (Figure 8e) and 87R (Figure 8f-h) plates show a similar pattern. Along with the DP skeletons, high stains the infused DP phases may shield the stored
strain energy in the W-particles, thereby retarding W-cleavage fracture. Dimple-like features of the DP phase inside the W particles supports this hypothesis (Figure 8h). Some WC local fracture mode was also observed for the 87R plate. However, the DP phase was mostly absent in these W-particles, consistent the lower toughness in the 74R plate.

Ongoing and Future Work

- The rolled 90W plates are highly anisotropic. Therefore, orientation dependent fracture toughness should be explored.
- The individual properties of the rolled W and DP phases will be explored by nanoindentation that will help to understand the global rolled W properties, and will be used as a modeling variable.
- ESBD with fine scanning steps, along with TEM, will further help to understand the toughness difference between 74 and 87R plates.
- Size-effects are expected and will be explored.
- A multi-mechanism toughening model will be developed.
- High temperature (600-800°C) fracture toughness properties of relatively larger (50mm x 12.5mm x 6.2mm) specimens should be performed in controlled environment.
- Thermal shock tests should be performed on these WHA's.

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Figure 8. The (a-d) Secondary electron SEM images are showing the fractographs for the 0, 62, 74 and 87R plates, respectively. The backscattered electron SEM image on 74R shows very minimal DP phase inside the W particles (e), while Figure f-g show a higher amount of DP phase infused inside the rolled W-particles for the 87R plate.

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4.2 SIMULATION OF MECHANICAL PROPERTIES OF TUNGSTEN COMPOSITES—L. M. Garrison, N. Jensen (Oak Ridge National Laboratory)

OBJECTIVE

The objective of this project is to perform a finite element analysis of a tensile test simulation of tungsten composites to understand and optimize their properties for use as plasma facing components in fusion reactors.

SUMMARY

As a first step toward modeling tungsten composites, simplified tensile tests of individual tungsten foils and composites were simulated. To realistically define the material properties required for modelling with ANSYS, experimentally measured stress-strain data from uniaxial tensile tests was imported. Strength, elasticity, and other relevant material properties were manually defined for tungsten, steel, and copper. The tensile frame contacts were simulated by modelling circular contact surfaces where the frame and specimen touched. One side of the contacts were selected to be fixed supports and a velocity of 0.1 mm/min in the axial direction was applied to the other. A finite element analysis of the uniaxial tensile test simulation solved for the stress, strain, and deformation in ANSYS. After the tensile simulation was completed, a follow-on simulation was done to simulate a layer breaking in the composite.

PROGRESS AND STATUS

Introduction

There are several types of tungsten composites being considered in the fusion community, including sintered, laminate, and tungsten-fiber reinforced materials. Many of these options have been successfully fabricated on a trial basis, but they have not been optimized. However, simulation of tungsten composites facilitates easier parameter modification, anticipates material properties and behavior, and identifies trends while avoiding the costs of time and resource intensive fabrication.

Of the tungsten composites considered in the fusion community, the laminate composite was chosen to model first. To properly model a laminate with such complex interface properties, individual tungsten, steel, and copper foils must be accurately simulated to reduce error in modelling the composite material. Foils of varying thicknesses can then be combined in ANSYS finite element analysis software to model laminates. Individual foils and a variety of laminates will be simulated in tensile tests to compare material and mechanical properties.

Results

The materials parameters used were listed in the last semi-annual report. The geometry is the SSJ tensile bar which is 16 mm long and 0.5 mm thick. The first type of composite modeled had three layers of tungsten alternating with two layers of steel, each 0.1 mm thick. Then, the composite layer thickness was varied as in Table 1. The tensile geometry is modeled in ANSYS and uses cylinders to approximate the tensile fixture contact on the shoulders of the samples to apply the force (Figure 1).

Number	of	tungsten	Number of steel layers	Tungsten	layer	Steel layer thickness
layers				thickness		
2			3	100 µm		100 µm
3			2	100 µm		100 µm
3			2	150 µm		25 µm
5			5	50 µm		50 µm

Table 1:	Tungsten	and steel	layer	configurations
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Figure 1. A 0.1 mm/min constant velocity was applied to the right circular contact pieces in the positive x direction with "A". A fixed support was applied across the faces of the left contact pieces marked with "B".

Typically, as a default, ANSYS outputs stress and strain values as the maximum experienced in any mesh element in the model. While this can be useful to understand the parameters across the sample, for the closest comparison to the experimental results, an average over the gauge cross-section was desired. This was implemented by using a plane at the center of the tensile bar, measuring the force at each element there and averaging across the cross-section area to achieve the engineering stress of the model (Figure 2) for comparison with the engineering stress measured experimentally.



Figure 2. Force measurement on the SSJ2 cross section.

Figure 3 shows the relationship between the percentage of tungsten in the composite and the ultimate strength. The data shows a linear relationship between the percent of tungsten and the ultimate strength of the composite.



Figure 3. Ultimate strength of different tungsten-steel composites.

After the different layer configurations were tested, a failure simulation was performed. To simulate artificial fracture, the ANSYS kill element command was used. The stress in each element was checked at each time step and the element was deleted if the stress was greater than a specified value. This simulated a crack suddenly propagating across one tungsten layer. The sudden deletion of a section was not intended as an accurate prediction of when fracture would occur, but it allowed study of how the composite would behave after one layer failed. The kill element command was also limited to a small sliver of material in the center of the outer tungsten layer, as seen in red in Figure 4.



Figure 4. The red section is the region that was deleted when the stress in the composite reached a specified value to simulate a fracture in one tungsten layer.

The equivalent stress of the sample was examined to see how the stress distribution changed after partial failure. Figure 5 shows that like the unfractured simulations, the tungsten layers sustained more stress than the steel layers. The middle tungsten layer experienced the bulk of the stress redistribution after the outer tungsten layer failed.



Figure 5. Equivalent stress distribution in W-steel composite with fractured tungsten layer.

4.3 MECHANICAL PROPERTIES OF TUNGSTEN IN THE PHENIX COLLABORATION IRRADIATION L.M. Garrison, J. R. Echols, N. Reid (Oak Ridge National Laboratory), T. Miyazawa (Tohoku University)

OBJECTIVE

The PHENIX collaboration tungsten irradiation aims to expand the database on neutron irradiation data for tungsten materials.

SUMMARY

The RB*19J capsule included over 20 varieties of tungsten and had three temperature zones, nominally 500, 800, and 1200°C. Tensile tests on all materials have been completed at the irradiation temperatures. Fracture surface imaging was completed in the scanning electron microscope (SEM). Hardness tests on all materials have been completed.

PROGRESS AND STATUS

Tensile tests of all materials have been completed at the irradiation temperatures, and SEM images of the fracture surfaces have been collected. The results of two materials are shown here, ALMT produced polycrystalline tungsten (Figure 1) and Goodfellow produced single crystal tungsten (Figure 2). For all conditions, the irradiated material is stronger and less ductile than the unirradiated sample tested at the same temperature. Both the polycrystalline W and the single crystal tungsten were brittle after irradiation to the middle dose and temperature, 776°C and 0.69 dpa for the polycrystalline and 830°C and 0.74 dpa for the single crystal.



Figure 1. Polycrystalline tungsten machined in the "A" orientation and tensile tested before and after irradiation at test temperatures of 500, 700, and 900°C.



Figure 2. Single crystal tungsten with <110> orientation along the tensile direction and tensile tested before and after irradiation at test temperatures of 500, 700, and 900°C.

The materials were microhardness tested with five intents averaged for each material. The results from the thermal neutron shielded PHENIX campaign are compared against the <110> single crystal tungsten that was previously irradiated without thermal neutron shielding in HFIR (Figure 3). Each data point of the shielded series represents a different type of unalloyed tungsten. The unshielded tungsten shows exponential increase in hardness with increasing dose and no dependence on irradiation temperature between 90 and 830° C. The thermal neutron shielded tungsten had similar hardness as the unshielded tungsten for samples irradiated to ~0.2 dpa. The thermal neutron shielded tungsten had noticeably lower hardness at ~0.7 dpa and higher doses. Interestingly, the thermal neutron shielded tungsten does have a dependence on irradiation temperature, with the material irradiated at the highest temperature having lower hardness than the material irradiated to similar doses and lower temperatures.



Figure 3. Microhardness of irradiated tungsten. All the unshielded tungsten is single crystal. Each data point of the shielded series is a different type of unalloyed tungsten (different grain sizes and manufacturers).

4.4 DAMAGE-MECHANISM INTERACTIONS AT THE PLASMA-MATERIALS INTERFACE—C. M. Parish, D. Morrall (Oak Ridge National Laboratory)

OBJECTIVE

The overarching objective of this work is to bridge the gap between the atomistic knowledge and models and the phenomenological materials science underlying the design, fabrication, and service of divertors and other plasma-facing materials for magnetic confinement fusion. Specifically, the influence of intrinsic defects (dislocations, grain boundaries) and extrinsic defects (ion- and neutron-irradiation damage, impurities) interactions with He and bubbles, in terms of nucleation sites, growth, trapping, and surface degradation, will be measured.

SUMMARY

This year we have explored both radiation damage and plasma-implantation effects. Radiation effects studies including continued analysis of neutron-irradiated tungsten, and plasma effects included analysis of the flux and fluence effects on the near-surface region of tungsten, and exploration of helium effects on the near-surface microstructure.

PROGRESS AND STATUS

First, neutron-irradiated tungsten specimens from the 19J gadolinium-shielded campaign were examined. Electron backscatter diffraction (EBSD) was used to examine the grain sizes of the polycrystalline specimens. Thanks to the new Oxford Symmetry EBSD + Tescan MIRA3 microscope at ORNL, unprecedented numbers of grains could be measured, giving excellent statistics for the evaluation of changes (or lack thereof) in the grain sizes. Indeed, having tens of thousands of grains for grain-size calculations now opens a new question of finding the correct statistical descriptions to determine when two distributions are the same or different, which is an open question.



Figure 1. The EBSD images (left) and measured grain sizes (right) for thick plate and rolled foil tungsten, irradiated to ~0.5 dpa at 500 or 800°C.

These observations are important, because eventual plasma-facing service behavior will depend significantly on the grain sizes, textures, and morphologies, and measurements as a function of irradiation parameters are necessary to understand the eventual plasma-facing behavior.

In addition to grain sizes, the precipitation and trans mutant microstructures were examined. The CT06 (thick plate, ~0.5 dpa 800°C) sample is shown in Figure 2. Here, a high density of voids is present, and X-ray mapping shows Re-enriched spherical volumes, consistent with solute clustering prior to precipitation of a second phase.

Data analytics of the EDS spectrum image, Figure 3, clearly shows the Re precipitates and Re segregation on the grain boundary.



🛾 70 nm

Figure 2. The STEM image (left) and X-ray maps (center, right) of the 800°C, ~0.5 dpa CT06, thick-plate tungsten from the Gd-shielded campaign. A high density of voids (black in HAADF) and Re-clusters (Re map) are visible.



Figure 3. Abundance maps and endmember spectra from the data in Figure 2, using a modification of independent component analysis. The red regions are the differences between endmembers #0 and #1 and are associated with Re L X-ray lines.

Several experimental campaigns are underway on plasma-materials interactions in collaboration with UC-San Diego. One campaign involves loading iso-fluence helium into the near-surface region of polycrystalline tungsten specimens at different temperatures in order to determine what effect a helium bubble "blockade" might have on fuel retention, and another involves iso-thermal loading of (001) single crystals to different fluences of helium.

Both campaigns are at low temperature (≤600°C) and low helium landing energy (~50 eV), so all the plasma-materials interactions are in the top ~15 nm of the specimens. However, FIB-based methods cause ~20-30 nm of damage in tungsten, so we have developed surface-specific FIB protocols to ensure the FIB preparation does not introduce excessive artifacts into the helium-implanted region. Figure 4 illustrates the FIB-deposited Pt cap, but with an electron-beam-deposited carbon cap beneath the Pt. This electron beam deposition provides a non-damaging sacrificial layer to prevent the gallium beam from destroying the features of interest during the Pt deposition.

This improved protocol is compared to the "standard" (incorrect) protocol in Figure 5, where significant Ga deposition and Ga/W intermixing is detected in the top ~15 nm of the specimen with the incorrect procedure, but with the improved protocol, the Ga and W layers are clearly separated and the He-induced damage is visible in the W layer's top few nanometers.



Figure 4. The SEM images of the tungsten specimen (left) with deposited sacrificial layers, and the FIB liftout with the electron-beam induced protective cap visible (right).



Figure 5. X-ray maps and STEM images of low-energy He-implanted tungsten prepared incorrectly (left) and correctly (right). The Ga and W layers are clearly differentiated in the correct procedure and intermixed in the incorrect.

Future Plans

Our collaborations with UC-San Diego and University of Illinois will continue. We will be performing advanced characterization on UIUC specimens and will possibly begin collaborating with Stony Brook University. The newly perfected FIB procedure is in use to prepare samples from the PMI campaigns. More FIB samples will be prepared from the 19J specimens for advanced STEM, as well.

4.5 INSITU MECHANICAL TESTING AND INTERFACIAL CHARACTERIZATION OF DUCTILE-PHASE TOUGHENED TUNGSTEN—James V. Haag IV, Mitsu Murayama (Virginia Tech), Matthew Olszta, Danny Edwards, Charles H. Henager Jr., Wahyu Setyawan (Pacific Northwest National Laboratory)

OBJECTIVE

The aim of this work is to study the mechanical behavior of a bi-phase W-Ni-Fe alloy subjected to a series of thermomechanical treatments. Hot-rolling to 87% thickness reduction fundamentally alters the microstructure of these alloys leading to a high degree of anisotropy, therefore adding a strong directionality component in their mechanical behavior. This treatment also increases the connectivity of the tungsten phase, leading to microcracking at W-W grain boundaries which dictates how the alloy accommodates plastic deformation. Combining SEM-based 3D volume reconstruction of a bi-phase W-Ni-Fe alloy before and after thermomechanical treatment with multidirectional and multiscale in-situ mechanical characterization techniques as well as transmission electron microscopy (TEM) interface analysis will provide much needed data for the validation or rejection of this material as a prime candidate for plasma facing material components (PFMCs) in fusion reactors.

SUMMARY

Tensile specimens of a hot-rolled W-Ni-Fe alloy were fabricated from three different orientations, i.e., TD, RD, and ND, via wire electrical discharge machining. These specimens were mechanically ground and polished for in-situ mechanical testing inside a scanning electron microscope (SEM). The samples were strained at a rate of $2 \cdot 10^{-5}$ s⁻¹ until failure. The microstructure response of the gauge region was video recorded during testing, allowing for observation of the deformation mechanics leading to failure and correlation with stress-strain results. The STEM-based electron diffraction mapping of the BCC W-rich phase and FCC Ni-Fe-W phase interfaces was conducted for crystallographic orientation and strain mapping analyses. Preliminary in-situ TEM mechanical testing was performed with video acquisition in bright field conditions to determine the strength of the dissimilar phase boundary. Further multiscale mechanical testing and orientation mapping is in process for greater statistical significance in the experimental analysis of this material.

PROGRESS AND STATUS

Introduction

Tungsten heavy alloys (WHAs) are gaining traction as alternatives to polycrystalline tungsten for plasma facing material components in fusion reactors due to their remarkable toughness [1-3]. Traditionally, WHAs consist of roughly spherical body centered cubic (BCC) W particles suspended in a face centered cubic (FCC) Ni-Fe-W matrix [4-5]. However, recent studies have been conducted on thermo mechanically treating these WHAs to tailor their microstructures into a form which exhibits both high strength and stiffness [6-7]. This aforementioned microstructure manipulation takes inspiration from nature, mimicking a 'brick-and-mortar' microstructure like those found in mollusks which exhibit a combination of high strength, stiffness, and toughness [8-9]. These tungsten-based biologically inspired composites are made through a combination of hot rolling and annealing steps until they resemble a series of hard stacked plates held together by a soft binder phase. The mechanical properties exhibited by these composites are a result not only of the physical properties of each constituent phase, but also of the phase directionality, continuity, and mechanical stability of the interphase boundary. Figure 1 shows a graphical illustration of the initial and final microstructures after this processing.



Figure 1. Hot rolling process for W-Ni-Fe. Industrially manufactured sample is top right; rolled sample is bottom left. Note the strong directionality present after hot rolling. All tensile bar orientations tested are shown.

An important factor in the characterization of these materials is that conventional 2D techniques do not provide enough understanding of the highly anisotropic phase distribution formed by thermomechanical treatment. As such, to properly characterize the microstructure, it is essential to see the material from a three-dimensional perspective. To understand the role of anisotropy from thermomechanical treatment in WHAs, the rolled material is mechanically tested in multiple directions, and the results are compared with bulk 3D reconstruction data to create a more robust framework for the description of the material and its behavior.

Experimental Procedure

Three different orientations of uniaxial tension specimens were milled as seen in Figure 1. The samples are defined by their directions relative to the processing directions, herein called the rolling direction (RD), transverse direction (TD), and normal direction (ND). Each sample was polished down to a mirror finish on both sides and tested at room temperature at a strain rate of $2 \cdot 10^{-5} \text{ s}^{-1}$ in uniaxial tension until failure in a Kammrath & Weiss micro tensile tester equipped with a 5kN load cell. This mechanical testing apparatus was placed inside an SEM operated at 30kV, so that video and physical testing data could be acquired

simultaneously. This allows for the comparison of observed deformation mechanisms with acquired stress and strain values. Video of the tensile tests show the initiation of discrete microcracks at W-W grain boundaries roughly perpendicular to the tensile direction starting at the yield point. These cracks are blunted by the soft Ni-Fe-W phase, and the number density of microcracks increases until a sufficient stress is reached to fracture the material, in the orientation shown in Figure 2 corresponding to ~625 MPa and 16% elongation. The composite also shows remarkable adherence between the dissimilar phases when fractured. Analysis of the fracture surface seen in Figure 2 reveals mixed inter- and intragranular fracture of the tungsten. The cleavage of tungsten grains is likely a result of sufficient stress localized at a single point to fracture the grain. Additional tensile tests are being conducted in triplicate on all three orientations of tensile specimens shown as well as on the unrolled sample for a comparative analysis of the mechanical behavior.



Figure 2. The SEM in-situ mechanical testing. (Top Left) SE image of ND-TD tensile specimen postfracture. (Bottom Left) Stress-strain graph of tensile test acquired during imaging. (Right) BSE image of fracture surface showing mixed mode failure in tungsten. Cleavage is shown with a red arrow, intergranular fracture is shown with a white arrow.

This in-situ testing data can be combined with a 3D reconstruction technique called mechanical serial sectioning to determine the effect of three-dimensional phase dispersion on observed mechanical properties. This technique is done by an iteration of imaging a sample surface and removing a known thickness from the surface by mechanical polishing. This process is repeated until a desired volume has been imaged. This process can be used to reconstruct volumes in excess of 1mm³. Figure 3 is a projection of a 250 x 250 x 250 μ m 3D reconstructed volume of both the industrially produced and rolled samples to show the drastically reduced connectivity of the Ni-Fe-W phase after processing. Failure of the unrolled

sample is dictated by the mean free path through the Ni-Fe-W phase from one edge to the other, while that cannot be the case in the rolled sample because of the discrete plate-like morphology of the soft phase.



Figure 3. The 3D Reconstruction showing only the Ni-Fe-W phase before hot rolling (left), and after hot rolling (right).

To elucidate the nature of the interphase boundary and its contribution to the overall behavior of the material it is necessary to move to the TEM length scale. Multiple sources have noted issues with the preparation of consistent and damage free tungsten and WHA TEM samples [10-11]. So, for additional thinning and removal of potential surface damage from focused ion beam (FIB) thinning, flash electropolishing was implemented. The setup conditions were based on information from references [11-12] and optimized for the setup shown in Figure 4. An electrolyte solution of 1% perchloric acid in a mixture of methanol and water, a 99.95% W foil cathode, 20V, 5ms pulses, and in an ice water bath were chosen as the best conditions for the given geometry, and a TEM image of the rolled WHA foil is shown in Figure 4. Samples were pulsed, then rinsed first with ethanol followed by reverse osmosis deionized (RODI) water after each step. This process was repeated until desired thinning was achieved. It should be noted that both phases

will not electropolish evenly due to differing electrochemical potentials between species, this step was only intended for further thinning and specimen preparation process induced damage removal.



Figure 4. (Top Left) Diagram of flash electropolishing setup connecting DC power supply to sample with timer and multi-meter in circuit to control pulse time and record maximum current. Sample is held by gold self-closing tweezers in a holder ensuring even spacing from cathode. Cathode is submerged in electrolyte which is held in cooling bath. Sample is placed in electrolyte, then pulsed to remove material. (Top Right) BF TEM image of sample after polishing. (Bottom) Image of setup in Virginia Tech lab space.

To observe the mechanical stability of the dissimilar phase boundary, a TEM foil of the rolled sample was prepared as seen in Figure 5. A FIB was used to mill an electron transparent window into the middle of the bar, acting as a notched specimen for tension testing. The sample was then deformed at a displacement rate of 20 nm/sec while in-situ video was recorded on a FEI Titan TEM operated at 300kV in TEM bright field mode. Figure 5 shows two still frames from the deformed region. It can be noted that the crack at the bottom middle of both frames has progressively widened, as well as generating a high density of dislocations in the interior of the Ni-Fe-W grain. A point of interest in this test is that there is little to no

dislocation generation and propagation in the vicinity of the interphase boundary. Previously acquired EDS line spectra at the same type of phase boundary reported a few nanometer wide chemical gradient at the on-edge interface, but this does not explain the lack of dislocation motion at the boundary. In-situ mechanical testing does show excellent adherence between the phases, even when tested in tension roughly perpendicular to the boundary, and also a region denuded of dislocation motion can be seen running parallel to the phase boundary in the Ni-Fe-W. Additional TEM in-situ mechanical tests will be conducted on different geometry boundaries to further explore the boundary effect on the toughening of tungsten heavy alloys.



Figure 5. The TEM in-situ mechanical test. (Top) Push-to-pull mechanical testing cartridge loaded with sample. Pins move closer together putting the sample in tension. Region enclosed in red box is milled to electron transparency with a Ga⁺ FIB. (Left) BF TEM image of phase boundary during tensile test. (Right) BF TEM image of same phase boundary region after ~10µm of cartridge displacement.

To more precisely determine the role of the phase boundary on the overall material behavior from a crystallographic perspective, a map of diffraction patterns was taken from an ion milled and flash electropolished sample of the rolled alloy. A phase boundary shown in the TEM micrograph of Figure 4 was chosen as the region of interest, and a series of diffraction patterns was taken every 1nm across a 50nm line perpendicular to the on-edge boundary. This was repeated 10 times along the boundary, represented by the different colored lines in Figure 6, creating a 50 x 10nm box analysis of the boundary. It can be confirmed that there is no presence of any intermediate phase between the BCC W and FCC Ni-Fe-W. It is also revealed that the orientation relationship between the two phases does not lie in a Kurdjumov-Sachs (K-S) or Nishiyama-Wasserman (N-W) type orientation, but instead a relationship close

to $BCC(\overline{101})\setminus FCC(\overline{151})$, $BCC[111]\setminus FCC[114]$. This map of diffraction patterns across the boundary also allows for the tracking of d-spacing changes in the region. This can be used to look at interfacial strain on each side of the boundary. Figure 6 plots the percent deviation from the expected lattice parameter on both sides of the boundary. Since the boundary is at x = 25nm, each point to the left of the boundary is indexed as W, while each point to the right is indexed as Ni-Fe-W. It can be noted that there is no noticeable change in the lattice parameter of the BCC W approaching the boundary, while the Ni-Fe-W deviates upwards of 3-4% from its expected value at the interface. The acquired EDS line scans validate this finding, showing that there is a chemical gradient transitioning from the W into the Ni-Fe-W at the boundary on the order of ~5-7 nm in width. This is the same result obtained from plotting the observed strain in the Ni-Fe-W lattice, and the observed lattice change in the Ni-Fe-W is likely a result of lattice expansion due to increased presence of tungsten in that region. A chemical gradient of W across the boundary would cause an expansion of the Ni-Fe-W lattice that linearly increases with increasing W concentration, following Vegard's Law [13]. This same analysis will be done on additional phase boundaries with different orientation relationships to track boundary width and directional strain as a result of lattice direction and crystal matching.



Figure 6. Diffraction mapping data from phase boundary. (Top Left) W [111] diffraction pattern on the left side of the boundary. (Bottom Left) Ni-Fe-W [114] diffraction pattern on the right side of the boundary. (Right) Calculated percent deviation from expected lattice parameter. The graph is plotted over a 50nm region in the x-direction. This x-distance is integrated over 10 different y-coordinates, represented by the different color lines. The expected lattice parameter is plotted as the W ($\overline{101}$) on the left side of the red line, and then Ni-Fe-W ($\overline{151}$) on the right side.

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4.6 PROGRESS IN THE ION IRRADIATION STUDY OF DUCTILE-PHASE TOUGHENED TUNGSTEN COMPOSITE—Weilin Jiang, Nicole Overman, Tamas Varga, Wahyu Setyawan, Charles H. Henager Jr. (Pacific Northwest National Laboratory)

OBJECTIVE

The aim of this experimental work is to investigate microstructures in ductile-phase toughened tungsten (DPT W) irradiated with He⁺ ions at an elevated temperature, especially at the W and NiFeW interfaces. In addition, helium retention in W and NiFeW grains will also be studied.

SUMMARY

A hot-rolled DPT W sample was characterized with various microscopy and spectroscopy methods, including backscatter electron (BSE) imaging, energy-dispersive X-ray spectroscopy (EDS), electron backscatter diffraction (EBSD) and x-ray diffraction (XRD) prior to 4 MeV He²⁺ ion irradiation. The EDS results show that W regions are 100% pure W and NiFeW regions contain 54.68 at.% Ni, 22.57 at.% Fe and 22.75 at.% W. The XRD data suggest that the average crystallite size for the W and NiFeW phases in the composite are 141 and 60 nm, respectively, and the weight percentage is 88 wt.% W and 12 wt.% NiFeW. Thus, the overall weight percentage in the composite is 90.8 wt.% W, 6.6 wt.% Ni and 2.6 wt.% Fe, consistent with the nominal composition of 90W-7Ni-3Fe for the DPT W. A DPT W sample has been irradiated with 4 MeV He²⁺ ions to 7.8×10^{17} He²⁺/cm² at 973 K. Our currently ongoing effort focuses the study of microstructures and He retention in the irradiated DPT W.

PROGRESS AND STATUS

Introduction

Tungsten possesses various favored properties for structural materials of fusion reactors, including high melting point (3695 K), good thermal conductivity (175 W/(m·K)), high resistance to material sputtering and chemical erosion, low swelling rate, low activation, and low tritium retention [1-6]. However, the material has low fracture toughness. Its brittle to ductile transition temperature (BDTT) increases with neutron irradiation damage [7]. Our current efforts have focused on the study of DPT W composites consisting of W and NiFeW phases [8]. Irradiation behavior of the W and NiFeW phases, including defect production rates and swelling, is expected to be different, which could impact the mechanical properties of the composite. It is important to study irradiation effects in DPT W, especially at the W and NiFeW interfaces. In addition, helium retention in W and NiFeW grains might be different, which could lead to a non-uniform helium distribution and gas bubble formation. We have been investigating the effects on the microstructure and He retention by using He⁺ ion irradiation of DPT W at an elevated temperature.

Experimental Procedure

The high-density DPT W composite (HD 17D) with a nominal composition of 90W-7Ni-3Fe was obtained from Mi-Tech [9,10]. The material was rolled at high temperatures (to 87% rolling reduction), cut into small samples, and polished on one side at the Pacific Northwest National Laboratory (PNNL) [8]. A DPT W sample (4 mm x 13 mm x 2 mm) was characterized prior to ion irradiation using a number of methods. Both BSE imaging and EDS were performed using a JEOL 7600 FESEM. The imaging magnification was 1000x at a working distance of 15.2 mm. To minimize the excitation volume for more accurate detection of the relatively small volume of NiFeW grains, the accelerating voltage for electrons was reduced to 5 kV. EBSD was also performed using a JEOL 7001F FESEM at an operating voltage of 20 kV. The EBSD data was collected at a specimen tilting angle of 70°. The scan was conducted over an area of 123 μ m × 92 μ m with a step size of 175 nm. Data analysis was accomplished using Oxford HKL Tango software. Unfortunately, there were no NiFeW phases in the database. Structural parameters for tungsten (ref. 229 with a = 0.31650 nm) and iron nickel (1/3) (ref. 225 with a = 0.35560 nm) were chosen for phase indexing. Symmetric 20- ω XRD was performed using a Philips X'Pert multipurpose diffractometer (MPD) with a



Figure 1. Quick Kinchin-Pease SRIM13 simulation results of 4 MeV He ion irradiation in tungsten for depth profiles of dose and He atoms. E_d : threshold displacement energy; E_b : lattice binding energy.

fixed Cu anode ($\lambda_{K\alpha}$ = 0.154 nm) operating at 45 kV and 40 mA. Whole pattern fitting (Rietvelt refinement) was performed to obtain the average crystallite size and weight percentage of the W and NiFeW phases using TOPAS-64 V6 software. Because the XRD database also does not contain any NiFeW phases, the best data fit was achieved using ICDD database PDF 00-004-0806(RDB) for W (Im-3m, a = 0.31648 nm) and ICSD database 103558 for FeNi₃ (Fm-3m, a = 0.35556 nm).

As described in our previous report [11], a portion of a DPT W sample was irradiated with a defocused 4 MeV He²⁺ beam to 7.8×10¹⁷ He²⁺/nm² at 973 K over an area of 3 mm × 3 mm. The irradiation was performed for a total of ~84 h using the 3.0 MV tandem ion accelerator facility located at the Texas A&M University. Quick Kinchin-Pease SRIM simulation [12] suggests that the maximum dose at the damage peak is 4.9 dpa at 6.16 µm and the maximum helium concentration is 24.6 at.% He at the depth of 6.24 µm, as shown in Figure 1. It should be noted that the actual He percentage in the profile could be significantly lower due to likely He outward diffusion and release during irradiation at 973 K.

Results

Table 1. Composition of Nil ever phase in DFT we determined by EDS									
Spectrum Label	Ni (wt.%)	Fe (wt.%)	W (wt.%)						
NiFeW1	54.91	22.61	22.48						
NiFeW2	55.83	23.46	20.71						
NiFeW3	53.38	21.81	24.81						
NiFeW4	55.92	22.91	21.18						
NiFeW5	53.35	22.09	24.56						
Average	54.68	22.57	22.75						
Standard Deviation	1.26	0.66	1.89						

Table 1. Composition of NiFeW phase in DPT W determined by EDS

Figure 2 shows a typical BSE image for a hot-rolled DPT W. The dark and lighter areas are NiFeW and W phases, respectively. Five areas from each of the phases (W1 – W5 and NiFeW1 – NiFeW5) were chosen for EDS analysis. The results from all the areas W1 – W5 are found to be 100% W, suggesting that the elongated and well interconnected particles due to rolling are pure W, which is consistent with a previous observation [8]. The results from areas NiFeW1 – NiFeW5 are given in Table 1. Also included in the table are the average weight percentages in the particles, showing 54.68 wt.% Ni, 22.57 wt.% Fe and 22.75 wt.% W with a standard deviation of less than 2 wt.% each. The small deviations are partly due to a



Figure 2. A BSE image for an as-rolled DPT W sample with areas indicated for EDS analyses. 5 keV electrons were chosen for the EDS to reduce excitation volume for probing small NiFeW grains in the DPT W.

small excitation volume from low electron energy (5 keV) for EDS analysis. It should be noted that trace oxygen (~1%) and carbon (~2%) were also present in the EDS spectra, but were removed as background contamination.

The EBSD pattern quality, phase and grain orientation maps of the as-rolled DPT W composite are shown in Figure 3, where the black areas are the unindexed regions that include grain boundaries and lattice distortions (strain). The unindexed, W and NiFeW regions occupy 1.325%, 89.14% and 9.54% areas, respectively. The mottled features inside the grains on the map may be attributed to the surface damage introduced either during polishing or rolling. From Figure 3, it is seen that the grain orientations in the out-of-plane (Z) view direction are randomly distributed without an obvious preferred orientation. Some of the individual grains within the grain show a color contrast, as shown in Figure 3. Thus, there may be some lattice stress in the grains.



Figure 3. EBSD pattern quality image, phase map and grain orientation map for an as-rolled DPT W sample.

Figure 4 shows the X-ray pattern for the DPT W sample. All the major diffraction peaks can be indexed, as indicated in the figure. The FeNi₃ is used for indexing NiFeW phase, as noted above. The whole pattern data fit suggests that the average crystallite size for the W and NiFeW phases are 141 and 60 nm, respectively. The average crystallite size is the average inter-planar distance between the adjacent planar defects that discontinue the lattice periodicity in a crystalline grain. In principle, crystallite size should not be greater than a grain. Typically, the smaller the average crystallite size, the higher the defect concentration in a crystalline grain. In addition, the data fit also indicates that the weight percentage is 88 wt.% W and 12 wt.% NiFeW in the DPT W composite. Considering the elemental weight percentages in



Figure 4. XRD pattern for an as-rolled DPT W sample.



Figure 5. A DPT W sample irradiated with 4 MeV He²⁺ ions to 7.8×10¹⁷ He²⁺/cm² at 973 K.

the NiFeW phase (Table 1), the overall weight percentage in the composite is 90.8 wt.% W, 6.6 wt.% Ni and 2.6 wt.% Fe, which is consistent with the manufacturer's technical specification for 90W-7Ni-3Fe.

A DPT W sample has been irradiated with 4 MeV He²⁺ ion to 7.8×10¹⁷ He²⁺/cm² at 973 K, which is shown in Figure 5. Our currently ongoing effort focuses on the preparation of thin foils using focused ion beam

(FIB) and flash polishing (FP) to remove the FIB damage on the specimen surface. The specimens will be examined using a Cs-aberration corrected scanning transmission electron microscope (STEM) equipped with electron energy loss spectroscopy (EELS) at PNNL. Microstructures in the W and NiFeW grains as well as their interfaces will be examined. Possible atomic intermixing at the W and W-Ni-Fe interfaces will be investigated. In addition, He retention in the W and NiFeW grains will be measured using EELS as a function of dose and He concentration. The results will be compared to the prediction from a density functional theory (DFT) calculation.

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4.7 MICROSTRUCTURAL CHARACTERIZATION AND MECHANICAL TESTING OF DUCTILE-PHASE TOUGHENED TUNGSTEN—Jing Wang, David Collins, Nicole R. Overman, Charles H. Henager Jr., Wahyu Setyawan (Pacific Northwest National Laboratory)

OBJECTIVE

The objective of the project is to understand the deformation behavior of ductile phase toughened W composites, such as W-Ni-Fe, for materials development for fusion reactor divertor and plasma-facing-component materials.

SUMMARY

This report summarizes the progress in mechanical testing and microstructural characterization for asreceived 95 wt.% W (95W) and 97 wt.% W (97W) W-Ni-Fe composites at PNNL. Two W-Ni-Fe composites were purchased from two different vendors (a total of four samples). Preliminary three-point bending and four-point bending tests were conducted on the samples, to provide direct comparison with previously tested 90W composites. In order to reduce challenges in integration with finite element (FE) model development and validation, the project determined to shift from bending to tensile tests for providing mechanical data. Micro-tensile specimens were fabricated from the as-received bulk samples via electrical discharge machining (EDM), and they were subsequently tested at room temperature under two different strain rates. As expected, the 95W composite samples exhibited better ductility than 97W samples. However, we found a significant difference in ductility between the two material vendors. Preliminary microstructural characterization on the samples were performed using scanning electron microscope (SEM). The general composition of W particles and Ni-Fe matrix, W particle size distribution, and the grain orientations were examined by electron dispersive x-ray spectrum (EDS) and electron back-scattering diffraction (EBSD). The result shows minimal microstructural and compositional variations between the material vendors.

PROGRESS AND STATUS

Introduction

Ductile phase toughening (DPT) is a fracture toughness improvement concept being used to develop tungsten-based composites for fusion reactor divertor and plasma facing materials. Tungsten is a promising candidate material for fusion reactor component applications due to its excellent high temperature strength, low sputtering rate, and high melting temperature [1, 2]. However, the potential application of tungsten as structural material is limited due to its low ductility, which could further degrade after irradiation [3]. Introducing a ductile phase for developing W composite could serve as an alternative route to overcome its limitations.

Previously, W-Cu composite materials have been investigated, and it is found that the ductile phase could form a bridge near the crack tip to enhance fracture toughness of the W-Cu composites [4, 5]. A finite element model was developed for understanding toughening mechanism (Dynamic Bridging Model), predicting load displacement curves and crack propagation patterns (Finite Element Continuum Model) [6, 7]. Although W-Cu composite is a good starting model system for exploration, it is not suitable for fusion reactor environment due to low melting point of the ductile phase Cu. An alternative system W-Ni-Fe composite was proposed and studied.

The effects of hot rolling were studied in the preceding fiscal years on the 90%W-7%Ni-3%Fe (wt.%, referred to as 90W) composite samples [8]. The as-received samples, in which W powders were embedded in a Ni-Fe matrix, were hot rolled at PNNL to 62%, 74% and 87% thickness reduction to resemble a lamellar structure. Single edge notch bend (SENB) bars were fabricated and tested using 4-point bend approach at PNNL. Finite element models developed for W composite were also calibrated and validated for the W-Ni-Fe composite material system. In FY19 and FY20, efforts to collect experimental data to understand the

effects of hot rolling and deformation behavior of W composite are continued. This report summarizes the progress on mechanical testing and microstructural characterization of the as received 95W and 97W composite materials, as listed in Table 1.

Vendor	Sample ID	Tungsten (wt%)
Plansee (Densimet [®])	D180	95
	D185	97
Mi-Tech	HD18D	95
	HD18.5	97

Table 1. W-Ni-Fe composite samples for mechanical testing and microstructural characterization

Experimental Procedure

Microstructural Characterization

Microstructural characterization on as-received samples were carried out using a JEOL 7600 field emission SEM at PNNL. Samples for characterization were first embedded in epoxy and the surface was polished to a 0.05 µm colloidal silica finish. A low angle backscatter electron (BSE) detector was utilized to examine the general microstructure at various locations using a 20-kV beam acceleration voltage. The chemical composition of W particle and Ni-Fe matrix in composite samples was examined using an Oxford Instruments Ultim Max 170 mm² EDS detector. The EBSD was performed using the Oxford Symmetry detector to acquire W grain size distributions and orientations. The results were processed and analyzed using the AZtec software package from the Oxford Instruments.

Mechanical Testing

Initially, mechanical testing was performed using the single-notch 3-point and 4-point bend geometries in order to be consistent with previous studies on 90W composite materials [9]. However, bending is difficult to model using finite element analysis due to large and unpredictable stress gradients as well as the pinsample contact points. To avoid such complexity, we choose tensile testing as the loading method. Current tests are focused on acquiring strain-stress curves, and testing using notched tensile samples will be performed to study fracture toughness in the future. Micro-tensile samples were fabricated via EDM from the as-received bulk materials into the geometry illustrated in Figure 1. Photos of the tensile test setup are shown in Figure 2.

For this set of tests, the samples were shoulder-loaded instead of pin-loaded to avoid tear-out at the pin hole as well as warping of the pins. The samples were tested at room temperature using an Instron 5582 servo-mechanical test frame equipped with an Epsilon ONE optical extensometer. The extensometer determines displacement by measuring the distance between contrasting color regions, in this case the white lines painted on the fixturing (as shown in Figure 2c)) and the sample. which was painted black. The strain of the gauge section is approximated by tracking the displacement of the white lines. Since only the change in distance of the gap in the fixturing is tracked using this technique, the effects of the compliance of the rest of the load train are eliminated. By normalizing the change in displacement to the nominal initial gauge length, an accurate estimation of the strain behavior of these materials can be obtained.

December 31, 2019







Tungsten Composite Tensile Sample

Units: Inches

Tolerances: ±0.002 unless otherwise stated Material: Tungsten composite, various types Parts Needed: 20 of each material

Figure 1. Tungsten particle composite tensile sample geometry.



Figure 2. a), b), c): the tensile testing setup from different viewing angles. Note the white stripes painted along the gap of the grips c).

While this test setup is suitable for obtaining estimations of strain behavior and demonstrating the qualitative differences between the materials, a more accurate measurement of strain is needed for FE model development and validation. However, the extensometer cannot directly track the change in gauge length of the specimen due to the fixturing obscuring the extensometer's view, as shown in Figure 2b). We are currently redesigning the fixturing to resolve this problem.

While the 95W and 97W materials are generally very hard and brittle, these composites showed some degree of ductile behavior in the preliminary 3-point bend tests. In addition, the observed displacement-stress relationships have shown strain-rate dependent behavior, where lower strain rates lead to apparent

increases in ductility. This effect is most likely associated with the relatively ductile matrix. In order to create a more accurate and comprehensive model, it is necessary to test these materials under a wide range of deformation rates. For these initial tests, two displacement rates, 10 and 0.1 microns per second (measured at the crosshead of the test frame), were applied. At each test speed, three tensile specimens of each material were tested. The slowest rate was chosen because it is approximately twice the displacement resolution of the test frame (~0.06 microns), which would mitigate the "stair step" pattern that is often seen in servo-mechanical test frames at extremely low displacement rates. The faster speed was set to two orders of magnitude higher in order to produce significantly contrasting data. To perform the tests, the samples were first preloaded to 10-20 N, then loaded at the prescribed displacement rate until failure. The resulting data was then smoothed, reduced to approximately 100 points, and plotted. Selected stress-strain curves are shown in the following section to compare the tensile behavior of each material at each loading speed.

Results

Microstructure

The W-Ni-Fe composite materials consist of two phases: nearly pure tungsten phase and a Ni-Fe-rich ductile phase as matrix. The BSE images for views of general microstructure of W-Ni-Fe composite samples are shown in Figure 3. The W particles in the 95W samples exhibit diameters on the range of tens micrometers, which are considerably smaller than those in 97W samples for materials despite vendor.



Figure 3. The BSE images of a) Plansee D180; b) Mi-Tech HD18D on the top row for 95W composite; and c) Plansee D185; d) HD18.5 on the bottom row for 97W composites. The scale given in d) applies to all other panels.

Figures 4-6 show inverse pole figure (IPF) maps, W phase grain sizes, and grain boundary characteristics from EBSD analysis of the W-Ni-Fe composite samples. The IPF maps show that the W-phase particles distribute randomly in the Ni-Fe matrix, and that the W particles are primarily single crystals. Slight grain rotations, colored as different orientations in IPF maps, are visible within some W particles, which was

below the angular limit defining a grain boundary, in this case 10°. Local variations in texture as shown are typically present when a material has undergone deformation. In this case the deformation could be present in the material intrinsically following material production. It is also plausible that the observed deformation could be primarily at the specimen surface, introduced as a polishing artifact. Or, both mentioned cases could be occurring simultaneously, because some of the local orientation shifts exhibit linearity that traverses grain boundaries, it is probable that some degree of surface damage is present.

Figure 5 shows the grain size distribution for the W phase. Because many of these W particles are single crystals, the grain size distribution is roughly equivalent to the particle size. Using the linear intercept method, results have shown that the W particles are about 50% larger in the 97W composite materials than in the 95W composite materials, which confirmed the qualitative observation in the BSE images. A preliminary analysis on the W-W grain boundary misorientation is shown in Figure 6. Most grain boundaries are high-angle grain boundaries (>15°) in all samples. A higher proportion of low angle grain boundaries with misorientation from 5-15° were detected in Plansee D180 (Figure 6a)). The identification of these low-angle angle grain boundaries coincides with the IPF textures within W-particles and is likely resulted from either deformation of the W particles that occurred during processing, or surface damage induced during polishing.



Figure 4. The EBSD Inverse Pole Figure (IPF) maps showing grain orientation in the out-of-plane (Z) direction. The top row corresponds to 95W specimens of the a) Plansee D180; b) Mi-Tech HD18D; and the bottom row corresponds to 97W samples c) Plansee D185; and d) Mi-Tech HD18.5.



Figure 5. Grain Size Distributions of the W phase. Top row corresponds to 95W: a) Plansee D180; b) Mi-Tech HD18D; and bottom row corresponds to 97W: c) Plansee D185; d) Mi-Tech HD18.5.



Figure 6. The EBSD Pattern Quality Maps for each of the evaluated specimens showing grain and phase boundary overlays for the 95W a) Plansee D180; b) Mi-Tech HD18D; and the 97W c) Plansee D185; d) Mi-Tech HD18.5. Red coloring indicates grain boundaries $\geq 5^{\circ}$, yellow $\geq 10^{\circ}$, and black $\geq 15^{\circ}$. White boundaries are indicative of phase boundaries.

Table 2 lists the compositions of Ni-Fe phase matrix and W phase particles in the W-Ni-Fe composite samples measured by EDS. Five different locations were selected from each phase, within each sample for improved statistics. The results indicate that the Ni-Fe phase matrix consists of ~25 wt% W, 53 wt% Ni and 22 wt% Fe, with a few percent variations from sample to sample, while the W phase particles are almost 100 wt% tungsten. A small difference in Fe concentration in Ni-Fe matrix can also be seen between Plansee samples and Mi-Tech samples (higher Fe content in the Plansee samples).

		D180		D185		HD18D			HD18.5				
Location	Spectra ID	w	Ni	Fe	w	Ni	Fe	w	Ni	Fe	w	Ni	Fe
Ni-Fe Matrix	#1	23.0	54.4	22.6	21.0	55.9	23.0	28.0	52.5	19.5	24.8	55.7	19.6
	#2	23.8	52.9	23.3	24.0	53.4	22.7	25.7	53.7	20.6	24.9	55.9	19.2
	#3	25.5	52.4	22.0	22.6	54.5	22.9	26.5	53.3	20.2	25.5	55.4	19.1
matrix	#4	24.2	53.2	22.7	24.4	53.4	22.2	25.1	54.5	20.4	24.5	55.8	19.7
	#5	24.7	53.1	22.3	23.6	53.9	22.5	25.1	55.3	19.6	27.0	54.6	18.4
Average (wt %)		24.2	53.2	22.6	23.1	54.2	22.7	26.1	53.8	20.1	25.3	55.5	19.2
Standard Deviation		1.0	0.7	0.5	1.3	1.1	0.3	1.2	1.1	0.5	1.0	0.5	0.5
W- Particle	#1	99.4	0.3	0.26	99.19	0.58	0.22	99.69	0.31	0	99.43	0.42	0.15
	#2	99.4	0.4	0	99.52	0.48	0	99.6	0.4	0	99.39	0.41	0.2
	#3	99.5	0.4	0	99.06	0.64	0.3	99.56	0.22	0.22	99.45	0.41	0.14
	#4	99.3	0.6	0	99.42	0.43	0.15	99.72	0.28	0	99.36	0.43	0.21
	#5	99.3	0.53	0.17	99.36	0.42	0.22	99.66	0.18	0.16	99.53	0.33	0.14
Average (wt %)		99.4	0.5	0.1*	99.3	0.5	0.2	99.6	0.3	0.1*	99.4	0.4	0.2
Standard Deviation		0.1	0.1	0.1*	0.2	0.1	0.1	0.1	0.1	0.1*	0.1	0.0	0.0

 Table 2. Composition (wt%) of Ni-Fe matrix and W particle measured at different points in W-Ni-Fe composite samples via EDS

NOTE: ¹Carbon was removed from the quantitative results shown above as carbon coating was utilized to ensure specimen conductivity. ²Oxygen was removed from the quantitative results of the W-Phase shown above as this finding was believed to be a result of surface oxidation. * Indicates statistical insignificance when averaged across five regions

Tensile Testing

Figure 7 shows representative stress-strain curves for the W-Ni-Fe composite samples tested at crosshead displacement rates of 10 μ m/s and 0.1 μ m/s. The ductility of each material is qualitatively consistent with the results of the preliminary 3-point and 4-point bend tests, with the Plansee D180 showing the highest ductility, the Mi-Tech HD18.5 showing the lowest ductility, and the Plansee D185 and Mi-Tech HD18D in between. Figure 7 clearly indicates that for each vendor the 95W composite is more ductile than the 97W composite. This result is expected as the ductile phase in this alloy (Ni-Fe matrix) has decreased in volume from the 95W to the 97W. Further, a second observed trend is the materials from Plansee are more ductile than the 97W counterparts, which is different from the general trend in a homogenous material where high tensile strength leads to a decrease in ductility. When tested at the slower displacement rate, the W-Ni-Fe composite samples exhibited decreases in apparent strength and increases in apparent ductility relative to the faster rate test, as expected.



Figure 7. Selected representative stress-strain curves for W-Ni-Fe composite samples tested at a) 10 μ m/s displacement rate; b) 0.1 μ m/s displacement rate.

The preliminary stress-strain curves and the 3-point and 4-point bending data (not shown here) both clearly show that the 97W samples are less ductile than the 95W samples from the same vendor, and both 95W and 97W materials from vendor Plansee are more ductile than those from Mi-Tech. The 95W material is more ductile because it contains a larger volume fraction of the Ni-Fe rich matrix phase. Multiple factors may have resulted in the increased ductility observed in the Plansee samples, including: 1) differences in the as-received material condition (annealed vs. unannealed); 2) a slightly reduced W-particle size of the Plansee samples; 3) variations in grain orientations within W particles; 4) differences in chemistry of the matrix (the matrix of the Plansee materials is approximately 54:20 Ni:Fe.); and 5) differences in the W-W grain boundary distribution, particularly the high-angle boundaries. Additional characterization of multiple areas from the as-received specimens following re-polishing could be mapped to verify if surface damage from polishing has impacted the results and improve statistics of the W-particle size. More importantly, post-mortem studies of the failed
tensile specimens will highlight any significant differences in deformation modes and failure locations that may have occurred. Another important factor is to consider the adhesion between the matrix and the tungsten particles, which would have a profound effect on the mechanical properties of these materials. Microhardness testing of the matrices of each material would be highly pertinent in this case, as would matrix-particle adhesion testing. The latter is very important for not only developing an accurate and a more physically based FE model, but also for determining the effects of the chemical differences of the matrices on the mechanical properties.

Future Work

The future work at PNNL includes testing and characterizing (before and after failure) the following DPT materials to provide data for model development and validation.

- 90W as-received material at room temperature.
- hot-rolled 90W materials (focused on the 87% rolling reduction, followed by the 62% and 74% reduction) at room temperature.
- Development of a new tensile grip design to allow for accurate strain data acquisition and the execution of additional tensile tests.
- Understand the source for the difference in ductility between materials from two vendors.
- Exploring the effects of matrix composition on mechanical properties.
- Extend the test and characterization to high temperatures to explore the limit of these composites.
- Extend the test and characterization to ion-beam irradiated composites to study the effect He, H, and atomic displacement damage.

Acknowledgements

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4.8 ELECTRICAL CONDUCTIVITY EVALUATION OF NEUTRON IRRADIATED TUNGSTEN MATERIALS FROM THE PHENIX CAMPAIGN—J. R. Echols, L. M. Garrison (Oak Ridge National Laboratory)

OBJECTIVE

The PHENIX collaboration is directed at expanding the database on neutron irradiation effects in tungsten materials. This task is evaluating the effects of irradiation on the electrical resistivity.

SUMMARY

After initial tests, the procedure for measuring electrical resistivity of 3mm discs was optimized and higher quality testing has been performed on a preliminary set of irradiated polycrystalline discs. The DPA level is shown not to likely be the only contributor to change in resistivity of PHENIX tungsten samples.

PROGRESS AND STATUS

The high thermal conductivity of tungsten is critical for its use in future fusion reactors and is one of the reasons tungsten is the leading candidate material for high heat flux regions of reactors. However, neutron irradiation in a reactor environment degrades the thermal conductivity of tungsten. To separate the phonon, electron, and trans mutant element contributions to tungsten thermal conductivity, electrical conductivity measurements need to be compared against thermal conductivity measurements. For the purposes of this report, electrical resistivity (the inverse of conductivity) is given. Ideally, resistivity (ρ) is defined as the resistance (R) of a uniform specimen of material multiplied by the cross-sectional area (A) and divided by the length (I) of the specimen, shown in Equation 1.

Equation 1.
$$\rho = R \frac{A}{I}$$

The US-Japan collaboration, PHENIX, has a goal of investigating tungsten and tungsten-based materials response to neutron irradiation for use in future fusion reactors. To this end, the PHENIX collaboration exposed over 1500 single crystal, polycrystalline, and W-Re alloy samples in the RB*19J irradiation capsule in the High Flux Isotope Reactor (HFIR) to doses of ~0.2-0.7 dpa. Nominal irradiation temperatures were 430-670, 740-960, and 880-1080°C. A gadolinium shield was included in the irradiation capsule to reduce the thermal neutron flux and therefore the rate of W to Re,Os transmutations.

The HFIR capsule geometry and radioactivity levels of the irradiated samples severely limit the size of the samples which can be used. Therefore, to measure the electrical conductivity of the irradiated samples, two different miniature electrical resistivity fixtures were designed and constructed: one for 3 mm diameter disks and one for SSJ tensile specimens. These fixtures are shown in Figure 1. The electrical resistance measurement system utilizes a Keithley Model 182 Sensitive Digital Voltmeter to measure the voltage drop across the inner lead, a Model 237 High Voltage Source Measure Unit which provides the current source, along with the custom fixtures. The system has been constructed, tested and calibrated against high purity W and Cu standards. Following calibration to a copper standard, unirradiated tungsten falls within 2% of literature values of electrical resistivity. Resistivity testing was performed at room temperature, between 20°C and 24°C. Resistivity values were then normalized to 20°C for comparison.

Necessary geometric/volumetric limitations from HFIR, and radioactivity levels of the post-irradiated samples severely limit the size of the samples which are used. Therefore, to test the electrical conductivity of the post-irradiation samples, a miniature electrical resistivity fixtures was designed and constructed for 3mm diameter disks. This fixture is shown in Figure 1.



Figure 1. Electrical resistivity testing apparatus. For each image, major gridlines are 1 cm apart. a) Disassembled 3 mm disk holder. Apparatus on left, example 3 mm disk and plug on right. b) Assembled 3 mm disk fixture. The plug is clamped to the apparatus to ensure proper contact with the pins. c) Simplified 3 mm disk circuit schematic. Circuit schematic specifies length (I), width (w), and thicknesses (t) used in resistivity calculation.

The testing apparatus utilizes a Keithley Model 182 Sensitive Digital Voltmeter and a Model 237 High Voltage Source Measure Unit along with the custom fixtures. The general test procedures for electrical resistivity testing on metals are given in ASTM B 193-87, Standard Test Method for Resistivity of Electrical Conductor Materials.

After noticing unexpectedly noisy data from initial tests, significant optimization of the system has been performed. This optimization involved standardizing the amount of compressive strain placed on the discs during testing, as compressive strain lowers a materials resistivity. Additional optimization involved the removal of presumed impurities on sample surfaces following irradiation and verification that resistivity values obtained are consistent regardless of sample thickness. Sample surfaces from the 880-1080°C zone is discolored by an opaque film. Measuring the resistivity of the samples before and after removing the film confirmed that this film would give inaccurately high results, as shown in Figure 2. Therefore, all samples from this zone must first be polished on one side before testing. Finally, the apparatus was confirmed to give accurate resistivity regardless of sample thickness.



Figure 2. Resistivity vs fast neutron fluence, for measurements taken between 20°C and 24°C and normalized for 20°C (left) and relationship between thickness and resistivity (right). The polished vs unpolished data shows the importance of removing the thin film apparent on samples from the 880-1080°C zone. The thickness data shows that following polishing, resistance of pure tungsten doesn't vary more than $2n\Omega m$.

Initial resistivity testing (Figure 3) with these optimized procedures and samples has been performed on thick plate, ITER grade, polycrystalline tungsten machined in various orientations relative to isotropic grains. The AT series samples are cut at 90 degrees relative to BT series samples.







Figure 3. Unirradiated grain structure (top) and resistivity vs DPA for AT and BT series materials (bottom). AT left and BT right. Irradiation temperature is given for all samples in the resistivity graphs and resistivity testing occurred between 20°C and 24°C and was normalized to 20°C.

Initial interpretation of the resistivity of these polycrystalline W samples would suggest that temperature and/or re-crystallation are contributing to an observed minimum of electrical resistivity in samples AT05 and BT05 which experienced approximately 0.68DPA each at around 750°C.

Future Plans

Testing will continue for polycrystalline W, as well as single crystal W, and W-Re alloys. Comparisons to irradiation temperature, transmutation, microstructure, thermal conductivity will be made in the future to investigate their relationship to the electrical resistivity.

5. MAGNETIC AND DIAGNOSTIC SYSTEM MATERIALS

No contributions this reporting period.

FUSION CORROSION AND COMPATIBILITY SCIENCE 6.

6.1 LIQUID METAL COMPATIBILITY IN FLOWING SYSTEMSJ. Jun, B. A. Pint (Oak Ridge National Laboratory)

OBJECTIVE

The goal of this task is to investigate the maximum Pb-Li temperature achievable in the dual coolant leadlithium (DCLL) blanket concept for the overall system efficiency. The FeCrAl alloys are potential candidates and a series of monometallic thermal convection loops (TCL) fabricated from a commercial FeCrAl alloy have been operated to assess the maximum use temperature where the alloy is compatible with flowing Pb-Li.

SUMMARY

A fourth monometallic TCL made from Kanthal alloy APMT (Fe-21Cr-5Al-3Mo) has completed 1000-hr operation with a maximum temperature of flowing Pb-Li at 700°C and a temperature gradient of 90°C. After exposure and cleaning, the specimens mass change was measured. Typical of prior experiments at lower temperatures, the specimens without pre-oxidation showed higher mass losses but most specimens showed a mass loss where the median values were similar to prior experiments. X-ray diffraction indicated that α - and γ -LiAlO₂ formed on the pre-oxidized and bare APMT, respectively.

PROGRESS AND STATUS

Introduction

An alumina surface layer is well-known to inhibit the dissolution of ferritic steels in Pb-17at.%Li and has the potential to raise the maximum operating temperature of the DCLL blanket concept for a demonstration reactor. Rather than depositing an alumina layer on a steel, a more robust solution is to aluminize the steel or explore Al-containing FeCrAl alloys to improve Pb-Li compatibility. The goal of this ongoing project has been to assess the maximum temperature where an Al-containing alloy could maintain good compatibility with flowing Pb-Li in a temperature gradient. A series of monometallic TCL experiments has been conducted using commercially available tubing material Kanthal alloy APMT (Advanced Power Metallurgy Tube) with each TCL having an increased maximum temperature as shown in Table 1 [1,2].

	550°C TCL [1]	600°C TCL [2]	650°C TCL [2]	700°C TCL	
ΔT/°C	115	85	120	85	
Flow velocity / cm/s	0.7	1.2	0.5	1.4	
Specimens in each leg	18 pre-oxidized SS3 2 as-received SS3	18 pre-oxidized SS3 2 as-received SS3	14 pre-oxidized SS3 4 as-received SS3 1 pre-oxid. coupon 1 as-rec. coupon	14 pre-oxidized SS3 4 as-received SS3 1 pre-oxidized coupon 1 as-received coupon	
Pre-oxidation	8 hr at 1050°C (18)	2 hr at 1100°C (2) 8 hr at 1050°C (14) 2 hr at 1000°C (2)	2 hr at 1000°C (13) 8 hr at 1050°C (2)	2 hr at 1000°C	

Table 1	 Details 	of the	1000 hr	monometallic	APMT TCI	_ experiments
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It was observed in the earlier TCL experiments that pre-oxidation of APMT to form an α -Al₂O₃ external scale significantly reduced the mass loss after exposure. However, at the highest exposure temperatures, the alumina was transforming to α -LiAlO₂, similar to prior observations in static Pb-Li [3]. Another issue was post exposure spallation of the alumina scale, which is a long-term concern for depleting Al from the alloy or coating. To minimize spallation, a shorter pre-oxidation of 2 hr at 1000°C was adopted for the most recent experiment compared to earlier experiments, Table 1. Also noted in Table 1, the majority of exposed specimens were tensile specimens to evaluate the post-exposure room temperature tensile properties.

Experimental Procedure

The fourth TCL experiment followed similar procedures as detailed previously [1,2]. Chains of 20 APMT specimens were exposed in the hot and cold legs of the TCL and the specimens are noted in Table 1. The coupons were 15 x 25 x 2 mm and the tensiles were 25mm long SS-3 type. Most of the specimens were pre-oxidized for 2 hr at 1000°C in laboratory air [2]. The APMT loop was pre-oxidized for 8 hr at 1050°C as part of the post-weld heat treatment procedure [1]. The temperatures measured during the experiment are shown in Figure 1. After the exposure, the PbLi was dumped into a stainless steel tank and the TCL was cleaned using the standard solution of acetic acid, ethanol and hydrogen peroxide [1]. After disassembling, it was discovered that the second specimen in the hot leg chain had broken and 3 other specimens were missing. An attempt is in progress to extract them from the dump tank. Specimens were weighed before and after exposure using a Mettler Toledo X205 balance. The coupons were analyzed using x-ray diffraction (XRD) and other characterization is in progress. Post-exposure room temperature tensile testing also is in progress.

Results

The mass change data are shown in Figure 2. The first specimen in the hot leg showed a much larger mass loss than expected. Unfortunately, no mass change data are available for the next 4 specimens in the chain. While there may be concern that the higher temperature caused more mass loss, particularly for the pre-oxidized specimens, an analysis of the median mass loss of pre-oxidized APMT specimens in the hot and cold leg chains did not show a significant overall effect of temperature, Figure 3. However, the APMT specimens exposed at ~680° and 700°C did show higher mass losses than any other pre-oxidized



Figure 1. Measured temperatures from the six TCL thermowells during the experiment.



Figure 2. Mass change of APMT samples as a function of estimated Pb-Li temperature.

specimens., Figures 1 and 3. Most of the specimens without pre-oxidation showed higher mass losses, consistent with the previous TCL experiments [1,2]. Presumably, during the time required to form a surface oxide layer (see below), more dissolution occurred compared to the pre-oxidized specimens.

Figures 4 and 5 show the XRD analysis results for the APMT coupon specimens without and with preoxidation, respectively. The larger coupon specimens are easier for XRD analysis than the small tensile specimens. Images of each specimen also are shown. Similar to prior results [2], the bare APMT specimens formed γ -LiAlO₂, while the alumina layer on the pre-oxidized specimens transformed to α -LiAlO₂.

Further characterization including room temperature tensile results and polished cross-sections is in progress and will be reported in subsequent reports. Future TCL experiments will not seek to explore higher temperatures. The next TCL experiment is planned to continue to use an APMT loop with a peak temperature of 700°C but expose specimens of ODS FeCrAI [4,5] and CVD SiC as more fusion relevant materials that have not been exposed to flowing Pb-Li. The APMT loop has been fabricated for this experiment and it is expected to start in the next quarter.



Figure 3. Median mass change values for preoxidized APMT specimens in all four TCL experiments.



Figure 4. The XRD results and photo images of bare APMT coupons exposed to Pb-Li in hot (top) and cold (bottom) legs. The estimated Pb-Li temperature and mass change for the coupons are in the inset tables.



Figure 5. The XRD results and photo images of pre-oxidized APMT coupons exposed to Pb-Li in hot (top) and cold (bottom) legs. The estimated Pb-Li temperature and mass change for the coupons are in the inset tables.

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7. ADVANCED MANUFACTURING

7.1 ODS FeCrAI PRODUCTION USING ADDITIVE MANUFACTURING WITH *INSITU* **OXIDATION**—Ty Austin, Steven J. Zinkle (University of Tennessee), Niyanth Sridharan (Lincoln Electric)

OBJECTIVE

The objective of this task is to explore the feasibility of producing oxide dispersion strengthened (ODS) FeCrAl + Mo + Y alloy parts using a laser directed energy deposition (DED) additive manufacturing (AM) process in an oxygen-rich environment, in order to achieve high-performance, geometrically complex ODS alloy components.

SUMMARY

The Fe - 12Cr - 6Al - 2Mo - 0.18Y (wt%) alloy powder was consolidated using a DED AM process in an oxygen-rich environment using varied build parameters to improve oxygen retention, reduce cracking, reduce porosity compared to our initial scoping experiments. In contrast to the previous experiments, this set of specimens were built under an inert argon atmosphere with an oxygen-rich welding gas stream. The oxygen content of as-built parts varied from approximately half of the as-received powder content to approximately half of a conventionally manufactured (powder metallurgy) ODS-FeCrAI. The specimen utilizing the slower raster speed maintained the highest oxygen absorption. This indicated that the oxygen absorption and retention, while being controlled by a solute trapping mechanism in previous work, was also heavily affected by the melt pool lifetime (a parameter limited by raster speed). For these experiments the laser was not pulsed to reduce thermal cycling and residual stress buildup. This led to a decrease in intergranular and trans granular cracking compared to previous experiments.

PROGRESS AND STATUS

The 20mm x 20mm x 20-30mm (height) sample cubes were built using a powder-blown, DED AM system built by DM3D Technology with gas atomized metal powder (Fe - 12Cr - 6Al - 2Mo - 0.18Y wt%) provided by ATI Powder Metals. To provide oxygen for oxide formation, the argon gas stream carrying the powder was blended with an Ar-10% O₂ gas mixture. This allows for the build chamber at large to be kept under a primarily inert atmosphere and provides better control over the oxygen absorption. During each build, the build chamber was kept at approximately 1 at% oxygen. A schematic of this process is shown in Figure 1 where diatomic oxygen provided in the carrier gas can diffuse directly into the hot melt pool or break down into monatomic oxygen before diffusing into the hot melt pool.



Figure 1: Schematic of DED AM process using in situ oxidation

For this set of experiments, samples were built under 6 different sets of conditions to examine how changing them would alter oxygen absorption. The following parameters were held constant: powder flow rate of 5 grams per minute, shaping gas flow rate of 12 liters per minute, cover gas flow rate of 12 liters per minute, nozzle-argon gas flow rate of 12 liters per minute, and a nozzle-helium gas flow rate of 2 liters per minute. The varied operating conditions are described in Table 1 below.

Build		Ar10O ₂ _1	Ar10O ₂ _2	Ar10O ₂ _3	Ar10O ₂ _4	Ar10O ₂ _5	Ar10O ₂ _6
Laser Power	· (W)	400	500	650	500	500	500
Gas Flow Rates	Carrier (Ar)	1.6	1.6	1.6	1.6	3.8	1.6
(L / min)	Carrier (Ar-O ₂)	4.4	4.4	4.4	4.4	2.2	4.4
Tool Speed (mn	n / min)	720	720	720	600	720	720
Pre-Heat Temperature (°C)		n/a	n/a	n/a	n/a	n/a	350
Final Layer Heig	ht (mm)	0.500	0.642	1.050	0.786	0.656	0.623

Table 1. Sample processing parameters

After fabrication, samples were taken from each specimen and sent to Dirats Laboratories for compositional analysis using inductively coupled plasma optical emission spectroscopy (ICP-OES) and combustion analysis using inert gas fusion (IGF). The compositional results are shown in Figure 2 along with the compositional results of two samples from the previous experiment (labeled 400WPNo and 400WC720) and a conventionally produced (ball milled, HIP'd/extruded) ODS-FeCrAI alloy included for reference [1]. The samples from the previous experiment were produced under an oxygen-rich atmosphere with a pure argon welding gas stream. They were produced with a laser power of 400W, tool speed of 720 millimeters per minute, no pre-heat, and with the laser pulsing for 0.08 seconds (400WPNo) or continuously running (400WC720). As shown in Figure 2, the major alloving element concentrations remain relatively stable across all samples. Oxygen absorption and retention were improved overall compared to the results from the first set of experiments. This increased oxygen absorption and retention were attributed to increased direct contact of oxygen with the melt pool when it is at its hottest temperature. In the previous set of experiments the oxygen contacted the melt pool after the laser (shrouded in an argon cone) had moved to the next portion of the sample. Oxygen absorption and retention were maximized in the sample with the slower tool speed, providing a 2.5x increase in oxygen content over the previous best sample and over a 6x increase in oxygen content over the as-received material. The same sample showed an oxygen content of approximately half of that produced with conventional manufacturing [1].



Figure 2. Compositional analysis of as-received powder, as-built specimens, and conventionally fabricated ODS FeCrAI [1]

In addition to compositional analysis, the specimens were cross-sectioned, mounted, and polished using conventional metallography techniques to prepare for microstructural characterization using a scanning electron microscope (SEM). The samples were also characterized using a Leica DM4000M optical microscope. The optical images of the specimens are shown in Figure 3.



Figure 3. Optical images of cross-section specimens (1-6). Top orientation is longitudinal (build direction to top of page). Bottom orientation is transverse (center of specimen to top of page).

As shown in the stitched optical images, significant cracking and porosity occurred in some of the as-built samples (particularly in sample 3). However, samples 2, 4, and 6 showed significant improvement in defect density – a measure of the negative spaces in the optical images such as cracks and porosity with respect to the whole metal sample – over specimens produced during the previous set of experiments, as summarized in Figure 4 below [2]. The sample with the highest oxygen absorption (sample 4) happens to also have the lowest defect density at approximately 0.69%, which is a particularly promising result. This can be contrasted with the previous experiment that exhibited a very high 8.68% defect density for the sample with the highest oxygen content.



Figure 4. Comparison of defect density of as-built specimens.

This decrease in defect density is attributed to a decrease in the cooling rate as a result of the laser moving more slowly across the sample. This results in slower or fewer thermal cycles that decreases the residual stress buildup in the sample and reduce cracking or hot tearing. It is also important to point out that the unusually high defect density for the $Ar10O_2_3$ sample is believed to be a result of poor powder flow. Powder flowability unexpectedly cut out during the production of the last few millimeters of this sample and resulted in excess amounts of energy being dumped into the part.

The improvement in build quality is further demonstrated in the Vickers hardness data. Using a LECO AMH 55 LM110 Series Automatic Hardness Tester, a 12 x 2 array of points were used to gather hardness data. Points were placed to avoid obvious pores or cracks on the surface, but subsurface defects were unavoidable. Overall the ranges and standard deviations of the hardness values shown in Figure 5 show a more consistent value compared to the two preliminary samples (400WPNo and 400WC720), indicating overall better build quality.



Figure 5. Comparison of sample build quality using Vickers hardness.

To better understand the cracking mechanisms at work, electron backscatter diffraction (EBSD) was performed using a JEOL 6500F FE-SEM equipped with an EDAX EBSD detector. The EBSD maps shown in Figure 6 demonstrated that intergranular and possibly trans granular cracking occurred in the Ar10O₂_4 build, the most promising sample thus far. Intergranular cracking indicates that stress buildup may be pulling the molten melt pool apart prior to complete solidification [3, 4]. Trans granular cracking implies the occurrence of large residual stresses on the solidified material exceeding the yield strength. We believe that the reduced tool speed contributed to the reduction in cracking by reducing the thermal cycling as discussed above. Future work will aim to further reduce this residual stress buildup and cracking.



Figure 6. Trans granular and intergranular cracking in the Ar10O₂_4 sample.

Future Plans

Transmission electron microscopy (TEM) and atom probe tomography (APT) specimens will be produced and analyzed to search for nanoscale oxide precipitation and to characterize the morphology and composition of the precipitation formed. Further experimentation will look at refining the operating parameters and possibly the alloy composition in order to reduce the crack formation and refine the precipitate distribution. These new samples will be characterized using electron microscopy and microhardness; tensile testing will be performed on samples that exhibit attractive microstructures.

Acknowledgements

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8. MECHANISMS AND ANALYSIS

8.1 INSITU SPECTROSCOPIC ELLIPSOMETRY FOR REAL-TIME CHARACTERIZATION OF THE EFFECTS OF HIGH-FLUX HELIUM PLASMAS ON TUNGSTEN SURFACES—R. D. Kolasinski, C. - S. Wong, A. Engel, J. A. Whaley (Sandia National Laboratories), F. I. Allen (University of California, Berkeley)

OBJECTIVE

The goal of this work is to demonstrate the use of spectroscopic ellipsometry for real-time characterization of tungsten surface morphology evolution during exposure to He plasmas. Insitu analysis of surfaces during high-flux plasma exposure represents a long-standing challenge in the study of plasma-material interactions. While state-of-the-art materials characterization can provide rich structural and composition detail during post-mortem analyses, in-situ diagnostics offer the possibility of probing dynamic effects. One of our main objectives here is to illustrate how the surface optical properties obtained with spectroscopic ellipsometry can be used to rigorously extract real-time structural information with nm resolution.

SUMMARY

Tungsten samples were exposed to He plasmas generated by an RF source ($\Gamma_i = 3.5 \times 10^{16}$ He cm⁻²s⁻¹, ion energy = 92 eV.) The range of exposure conditions selected here is conducive to the growth of near-surface He bubbles, and at higher fluence, the formation of W nano tendrils ranging between 50 – 100 nm in diameter. The evolution of these surface features was probed using a fixed-angle ellipsometer (280 – 1000 nm wavelength range) with direct line-of-sight to the sample. Over the parameter space explored here, changes in the two angles (ψ , δ) that define the polarization of the reflected light followed a distinct trajectory with increasing plasma fluence. Ex-situ ellipsometry of 22 additional tungsten specimens tested at a wide range of plasma fluences and temperatures mapped onto these in-situ results well. We used helium ion microscopy and focused ion beam profiling to provide a direct calibration of the ellipsometry measurements. Our results indicate that for a reproducible process such as the growth helium-induced surface morphologies, ellipsometry is a practical in-situ diagnostic to study how fusion plasmas modify materials. To study more general effects of plasmas on surfaces, including co-deposition and sputtering, different approaches to modelling the optical properties of the exposed surfaces are also considered.

INTRODUCTION

How intense fluxes of low-energy species (e.g. shallowly implanted hydrogen isotopes and helium) reconfigure the structure of surfaces is a topic of great importance in magnetically confined fusion plasmas. The plasma-facing surfaces of a fusion reactor will be exposed to a combination of high-flux deuterium-tritium plasmas, high-energy fusion products, as well as impurities (including artificially introduced species to promote radiative cooling of the scrape-off layer.) These energetic particles will continually reconfigure the exposed surfaces. While plasma-induced microstructures have been studied widely, how these structures evolve during plasma exposure from atomic-scale defects in initially pristine surfaces is much less well understood. Much of the knowledge gap arises from the difficulty associated with deciphering the large number of coupled surface processes that occur at the plasma-exposed surface. In addition, the difficult nature of characterization presents a formidable obstacle: while most conventional microscopy tools and surface analysis techniques work well in tightly-controlled ultra-high vacuum (UHV) environments, they are generally not compatible with the high pressures and magnetic fields encountered in a plasma environment. The time-dependent nature of the underlying mechanisms consequently has not been characterized directly and has contributed to a reliance on post-mortem analysis.

Insitu diagnostics involve instrumentation that is directly attached to the plasma chamber and monitors changes to the surface. An advantage of this approach is that it can capture dynamic effects that occur only during plasma exposure. Spectroscopic ellipsometry provides an intriguing alternative for quantifying changes in surface structure in-situ. It is an established technique for measuring the thickness of oxides grown on Si substrates and provides sensitivity to nm-sized roughening of surfaces. Commercial systems are available and have been adapted to in-situ applications such as monitoring thin film growth. The feasibility of extending this approach to larger scale linear plasma device was explored by Bastasz and co-workers [1] where a single wavelength ellipsometer was attached to

PISCES-B linear plasma device and used to measure the erosion rate of a SiO₂ film on a Si substrate during D_2^+ plasma bombardment. Herein, many of the practical challenges associated with insitu ellipsometry, including alignment and calibration, were identified.

In the present study, our goal is to extend ellipsometry to study more complex surface morphologies produced by He plasma exposure. A summary of the relevant conditions appears in a recent review by Petti [2]. A complete model of the growth responsible for the complex structures produced by He plasmas has not yet been developed, though considerable progress has been made in simulating the initial stages of this process.

PROGRESS AND STATUS

The plasma exposure chamber used in this study is described in two recent publications [3,4]. For this work, we operated the source with an applied axial magnetic field of 185 G, RF power of 250 W, and He neutral pressure of 0.65 Pa (5 mTorr.) The coil directs electromagnetic waves along its axis and ionizes the plasma with a non-resonant process (frequency: 420 MHz.) A Langmuir single probe attached to a linear translation stage allows measurement of the plasma parameters at different radial points within the plasma column. Of most importance here is the incident ion flux, 3.5×10^{16} cm⁻²s⁻¹.

For in-situ measurements, we attached a spectroscopic ellipsometer (J. A. Woollam, 280 - 1000 nm wavelength range) directly to the RF-plasma source vacuum chamber. Both the ellipsometer light source and detector were mounted to adjustable gimbals and aligned to a 70° angle with respect to the surface normal of the mounted specimen. All specimens were fabricated from polycrystalline tungsten (ITER-grade, Allied Materials Corporation) and mechanically polished to an RMS surface finish of 10 nm. The spot size from the beam of the ellipsometer required using 25 mm diameter specimens (2 mm thick) for in-situ analysis. For ex-situ analysis, smaller 6 mm diameter buttons were used for convenience. After polishing, all samples were heated to 900 °C in an ultra-high vacuum (UHV) furnace (< 10^{-7} Torr) over 30 min. and held at this temperature for 1 hr. Prior electron backscatter diffraction measurements on these materials indicate that grain size ranges between $10 - 20 \,\mu$ m across. Following plasma exposure, all specimens were imaged using a helium microscopy (Zeiss nanoFab) and atomic force microscopy.



Figure 1. Tungsten sample surfaces showing roughening and nano-tendril formation following high-flux helium plasma exposure at varying temperatures. For each exposure temperature, a "top" view perspective obtained using helium ion microscopy is shown. In addition, FIB cross sections are shown for T \ge 658 °C, with the estimated thickness of the nanostructure indicated. Plasma exposure conditions: $n_{\text{pl}} = 7 \times 10^{16} \text{ m}^{-3}$; $T_{\text{e}} = 11.5 \text{ eV}$; $E_{\text{ion}} = 92 \text{ eV}$; $\Gamma_{\text{ion}} = 8.5 \times 10^{20} \text{ m}^{-2} \text{ s}^{-1}$; $\Phi_{\text{ion}} = 1.53 \times 10^{25} \text{ m}^{-2}$.

To provide a suitable data set for exsitu analysis, a series of nine tungsten samples were each heated to a constant temperature between 472 - 937 °C. They were then exposed to He plasmas for 5 hrs, corresponding to a total ion fluence of 6.3×10^{24} He m⁻²s⁻¹. This set of exposure conditions produced the surface morphologies depicted by the sequence of HIM images in Figure 1. In each case, a top view of the surface is shown, along with a cross section created by focused ion beam profiling below. For the samples exposed at temperatures of 472 °C and 546 °C, the surface is covered with ripples roughly 20 nm in width, with small pits associated with ruptured bubbles just beneath the surface. At progressively higher temperatures, these ripples become more pronounced, and by 786 °C, 50 nm diameter tendrils begin to protrude. This tendril layer grows more quickly at higher exposure temperatures, with a thickness of 900 nm achieved at 937 °C following the 5 hr exposure time. This corresponds to an average growth rate of 3 nm/min.

Following plasma exposure, we characterized each specimen using exsitu ellipsometry. Spectra were acquired over the full wavelength range of our instrument (280 nm – 1000 nm.) To aid in interpreting the ellipsometry measurements, it is useful to consider how the two angles that define the polarization state of the reflected light (ψ , δ) change with the exposure conditions. These changes are typically rendered in the form of trajectories (ψ , δ) space, as shown by the examples in Figure 2. Here we have selected wavelengths in the UV (245 nm), visible (624 nm) and IR (999 mm) range. The blue markers indicate post-mortem measurements obtained for He-exposed W surfaces. The details of each data set are indicated in Table 1. The numbers by each marker indicate the thickness of the nanostructure layer, as measured by focused ion beam profiling. Despite the wide range of exposure conditions considered here, it is striking that the ex-situ data collapse onto a single curve. This is consistent with a nanostructure growth process that is reproducible and suggests that the fundamental mechanisms underlying the nanostructure growth remain the same within the range of parameter space explored here. It also indicates that the main effect of temperature is to accelerate or decelerate this growth process.

The red curves included each plot indicate in-situ measurements obtained during three exposures of different durations (ranging between 1-5 hrs), with each at a sample temperature of 900 °C. In all cases, the insitu data maps onto the post-mortem results well. While the data obtained at IR wavelengths follow a consistent trend toward higher ψ with increasing nanostructure thickness, a reversal of this behavior is observed at approximately 400 nm in the UV and visible wavelengths. We believe this is due to enhanced depolarization occurring once the nanostructure layer reaches a thickness comparable to the wavelength of light.



Figure 2. (ψ, δ) trajectory plots for W surfaces exposed to varying He plasma conditions. The symbols indicate data compiled exsitu measurements for individual specimens, whereas the solid lines correspond to insitu measurements. Data in the ultraviolet (245 nm), visible (624 nm) and infrared (999 nm) are shown. For the temperature dependence series, the thickness of the nanostructure layer is indicated in nm by the labels next to each marker.

data set	# of samples	summary
A	9	Sample temperature varied between 472 – 937 °C. Exposure conditions: E_{ion} = 92 eV; Γ_{ion} = 8.5×10 ²⁰ m ⁻² s ⁻¹ ; Φ_{ion} = 1.53×10 ²⁵ m ⁻² . Results reported in Ref. [3].
В	6	Ion fluence varied between $\Phi_{ion} = 5.0 \times 10^{23} - 1.2 \times 10^{25} \text{ m}^{-2}$. Exposure conditions: $T_{sample} = 840 \text{ °C}$; $E_{ion} = 92 \text{ eV}$; $\Gamma_{ion} = 8.5 \times 10^{20} \text{ m}^{-2} \text{ s}^{-1}$
С	2	Exposure conditions: $T_{sample} = 900 \text{ °C}$; $E_{ion} = 92 \text{ eV}$; $\Gamma_{ion} = 8.5 \times 10^{20} \text{ m}^{-2} \text{ s}^{-1}$, $\Phi_{ion} = 1.53 \times 10^{25} \text{ m}^{-2}$.
D	5	Exposure conditions: $T_{sample} = 840 \text{ °C}$; $E_{ion} = 90 \text{ eV}$; $\Gamma_{ion} = 4.0 \times 10^{20} \text{ m}^{-2} \text{ s}^{-1}$; $\Phi_{ion} = 8.6 \times 10^{24} \text{ m}^{-2}$. Results reported in Ref. [4].

Table 1. Summary of exsitu data sets for low energy He⁺ exposure of W surfaces

Summary and Outlook

With many of the practical aspects of the use of in-situ ellipsometry on linear plasma devices demonstrated, our forthcoming work focuses on extending this diagnostic to other problems of interest for plasma-material interactions. This includes studies of erosion and redeposition under higher energy (up to 1000 eV) ion bombardment. In addition, we are working to model the optical properties of the plasma-modified W surfaces, which is crucial information needed for interpreting the experimental data. In the forthcoming months, we are also planning to continue our collaboration with SciDAC program led by Brian Wirth to more broadly study the effects of He plasmas on W surfaces, with an emphasis on the early stages of W nanostructure formation.

From a practical standpoint, our work did not uncover any major obstacles to applying ellisometry to other linear plasma devices for in-situ measurements. The main constraint is the need for direct optical line of sight access to the sample. If this is not possible, the use of mirrors or other optical components would be required, as well as calibration to account for changes in the ellipsometric angles due to reflections. While the relatively modest magnetic fields present in our RF-plasma source did pose any issues for our ellipsometer, the higher fields in some larger plasma devices may affect the operation of motorized components. These effects usually can be mitigated through shielding. Consistency of surface preparation is also crucial.

Implementation of ellipsometry on a large plasma device such as a tokamak may also be feasible, though several significant hurdles must first be overcome. Optical access to the surfaces of interest will be an even greater challenge as available port space with the proper viewing angles would be limited and potentially spaced far apart. This makes maintaining optical alignment more challenging. In addition, device operation would preclude access to the ellipsometer, and as a result alignment, calibration, and measurements would need to be automated for this technique to be practical in a tokamak environment.

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8.2 SURFACE ANALYSIS AND HYDROGEN PERMATION MEASUREMENTS FOR SUPER-PERMEATING MEMBRANE MATERIALS—C. S. Wong, R. D. Kolasinski, J. A. Whaley (Sandia National Laboratories)

OBJECTIVE

The goal of this work is to characterize hydrogen-surface interactions and surface-to-bulk transport mechanisms associated with super-permeable membranes. On the upstream side of a super-permeable membrane, a low energy plasma is used to implant hydrogen ions beneath a thin oxide layer (potentially as thin as 1 monolayer.) The integrity of the oxide layer is critical as it must be relied upon to prevent hydrogen recombination and release. The opposite situation is true at the downstream surface, where fast recombination is needed to ensure adequate throughput of hydrogen gas. The goal of their relevance to super-permeation, we focus our efforts on understanding the effects on Group V metals (Nb, V, and Ta.) We will use a combination of surface analysis techniques and hydrogen permeation measurements to decipher the relevant mechanisms.

SUMMARY

In this report, we summarize preliminary surface characterization results for Nb surfaces, using low energy ion scattering, direct recoil spectrometry, and Auger electron spectroscopy. While most surface analysis tools cannot detect hydrogen, the low energy ion beam techniques described here are among the few techniques that are directly sensitive to it. For this study, we examined chemisorption using both molecular and atomic hydrogen (using an heated tungsten capillary to dissociate the hydrogen.) To complement these results, we have been performing ex-situ spectroscopic ellipsometry as a means of detecting the surface oxide.

PROGRESS AND STATUS

Surface Characterization

We characterized a clean reference niobium surface using low energy ion scattering (LEIS) techniques and Auger electron spectroscopy (AES). For these measurements, a 7-mm diameter polycrystalline niobium foil sample was prepared. The sample surface was first cleaned with successive ultrasonic baths of methanol and isopropanol, followed by rinsing in deionized water. Afterward the Nb sample was transferred into our ultra-high vacuum system (5×10⁻¹⁰ Torr for LEIS and 6×10⁻⁹ Torr for AES), where it was then sputtered cleaned using the 3 keV Ne⁺.

Our LEIS and direct recoil spectroscopy (DRS) measurements for the niobium sample were obtained with an angle-resolved ion energy spectrometer (ARIES). The experimental apparatus for ARIES has been described in detail in several references [1–6], so we provide just a short summary here. A mass separated 3 keV Ne⁺ beam was produced using a Colutron source. The beam was shaped and then steered with ion optics through an aperture into the primary measurement chamber. The chamber had a base pressure of 5×10^{-10} Torr, primarily due to residual H2(g). During operation of the ion beam, the pressure of the measurement chamber was maintained at 3×10^{-8} Torr by differential pumping. Hydrogen dosing of the sample was performed by introducing either atomic or molecular hydrogen into the chamber through a leak valve, until the chamber pressure increased to 2×10^{-7} Torr.

lon energy spectra of the niobium surface for the different three hydrogen environments are presented in Figure 1 for angles of incidence in the range $69^{\circ} \le \alpha \le 84^{\circ}$. We adopt the standard notation that X(QS) indicates a detected Ne+ that has undergone quasi-single scattering from a surface atom of species X, while X(R) indicates a detected X+ that has been recoiled by the incident Ne+. These spectra reveal that the surface composition is primarily niobium and hydrogen, with trace amounts of oxygen. As expected, the hydrogen peak increases when either molecular or atomic hydrogen are introduced into the chamber. However, it is notable that the hydrogen peak is identical in both cases. This indicates that molecular hydrogen is readily dissociated and adsorbed onto the niobium.

The ion energy spectra also reveal finer details. Firstly, the presence of hydrogen alters the shape of the Nb peaks. When hydrogen is added to the surface, the primary Nb(QS) peak undergoes a slight shift to lower energies, likely due to blocking and shadowing by the hydrogen. Secondly, the Nb(DS) peak is found to be suppressed by increasing the surface hydrogen. The Nb(DS) peak, which sits on the high energy shoulder of the Nb(QS) peak, corresponds to Ne+ with trajectories that have undergone two θ/2-collisions with Nb atoms. A final and less critical detail is the increase in scattered and recoiled oxygen for the atomic hydrogen dosing as compared to the molecular hydrogen dosing. This likely was caused by out gassing from the heated capillary used to dissociate the molecular hydrogen. To ensure that the peaks in the ion energy spectra are being attributed to the correct scattering and recoil processes, we constructed a scattering circles map, as described in Reference 1. This map, which is presented in Figure 2, was generated by obtaining ion energy spectra for scattering angles in the range $20^{\circ} \le \theta \le 90^{\circ}$, while α was fixed at 84°. The radial coordinate of the map corresponds to the square root of the normalized ion energy, while the polar coordinate corresponds to the scattering angle θ . Lighter regions in the map correspond to larger scattering and recoil intensities. These lighter regions are expected to trace out circles, which have been computed and overlaid on the map. The strong agreement between the overlaid circles and the underlying scattering and recoil intensity profile suggest that scattering and recoiling of the Nb, O, and H is sufficient to describe the obtained spectra.

Lastly, we performed Auger electron spectroscopy on the niobium sample. The AES complements LEIS and DRS, as it has higher sensitivity to other surface impurities, in particular, carbon. The obtained spectrum, shown in Figure 3, has niobium peaks that are consistent with the literature. The spectrum also reveals the presence of oxygen and carbon, which indicate that perhaps more substantial sputter cleaning is required to fully remove adsorbates from the surface.



Figure 1. Ion energy specta for 3 keV Ne⁺→Nb at angles of incidence $69^{\circ} \le \alpha \le 84^{\circ}$. These spectra were taken for the sample in three different hydrogen environments: residual hydrogen (black squares), H₂(g) dosing (red circles), and atomic H dosing (green triangles). Most prominent are the Nb(QS) and H(R) peaks, which correspond to Ne⁺ scattering off a Nb atom and H+ recoiled by an incident Ne⁺, respectively.



Figure 2. Scattering circle for the niobium sample for residual H conditions. The angle of incidence for this measurement was $\alpha = 84^{\circ}$. These data provide confidence in attributing the scattering and recoil peaks in Figure1 to niobium, hydrogen, and oxygen.



Figure 3. Auger electron spectroscopy of the clean niobium sample after sputter cleaning. The positions of the expected Auger peaks are given by the colored dotted lines (red for niobium, green for carbon, and blue for oxygen). The niobium peaks align extremely well with those from the literature, while the presence of oxygen and carbon were also detected.

SUMMARY

To build upon these preliminary results, additional surface characterization will be performed in a more representative plasma environment. We are working to set up in-situ spectroscopic ellipsometry measurements to study oxide sputtering and growth during low energy D₂ plasma exposure. Spectroscopic ellipsometry relies on reflecting a beam of elliptically polarized light from a surface. A detector is then used to determine how the polarization state of the reflected light has changed. This general approach can provide nanometer resolution measurements of oxide growth on semiconductor surfaces. We recently had success in developing a similar system for examination of tungsten nanostructure growth upon exposure to He plasmas. Applying this same approach to Nb, V, and Ta surfaces could provide a means of directly determining the oxide thickness as a function of plasma fluence, providing key information needed to assess the robustness of the oxide layer.

In addition, we have been working with the other national laboratories on a hydrogen permeation benchmarking exercise. Four labs agreed to participate in the effort, including Idaho National Laboratory (INL), Savannah River (SRNL), Oak Ridge (ORNL), as well as Sandia National Laboratories. In addition, Pacific Northwest National Laboratory and Los Alamos agreed to participate in an advisory capacity. As model system for comparison between laboratories, we will start with polycrystalline Ni. To begin with, Ni does not form a hydride and has a relatively high permeability. In addition, chemisorbed O is reduced in a H environment. This mitigates concerns about oxide growth affecting dissociation and recombination at the surface. Furthermore, there seems to be reasonable consistency in the literature database for H diffusion and permeation in Ni over a wide temperature range. To ensure that we are working within conditions that would be accessible to most instruments, will consider a temperature

range of 100 - 500 °C to start. Post-mortem depth profiling using Auger / X-ray photoelectron spectroscopy will be used to quantify the thickness of any oxides that have grown on the membrane surfaces.

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MODELING PROCESSES IN FUSION SYSTEM MATERIALS 9.

9.1 EFFECT OF CONFINEMENT OF SIA CLUSTER DIFFUSION BY IMPURITIES ON RADIATION DEFECT ACCUMULATION IN TUNGSTEN—Giridhar Nandipati, Wahyu Setyawan, Kenneth J. Roche, Richard J. Kurtz (Pacific Northwest National Laboratory), Brian D. Wirth (University of Tennessee)

Extended Abstract: Full manuscript will be submitted to a peer-reviewed journal.

It is well-known that adding certain impurities and minor alloying elements can improve the radiation resistance of polycrystalline materials [1-16]. The improvement is most pronounced in the temperature range of 0.3-0.55 T_m (T_m is the absolute melting temperature) [1, 2], where both vacancies and interstitials are mobile, and void swelling can occur. Impurities and alloy elements can reduce void swelling by acting as point-defect traps, thereby enhancing intragranular recombination between vacancies and self-interstitial atoms (SIAs). While both SIA and vacancy trapping are known to improve radiation resistance, SIA clusters are trapped more readily than vacancies because of their much more extended strain field interactions and faster diffusion. Therefore, the enhanced recombination is usually attributed to the decreased effective mobility of SIA due to the trapping. In other words, the role of vacancy diffusion is less known. Besides, in cases where SIA is only temporarily trapped, recombination may occur within the lattice and at the trapping sites, and the knowledge of which one is more prevalent is also less known. Note that, at elevated temperatures, depending on the binding energy between SIA clusters and the traps, SIA clusters may trap and de-trap (detach) numerous times before they recombine or are absorbed at defect sinks, such as dislocations, grain boundaries or free surfaces.

In tungsten, SIA clusters of all sizes glide one-dimensionally (1D) along their Burgers vector direction, and a change in their 1D diffusion direction requires overcoming of an activation barrier (rotation barrier). In pure tungsten, the energy barrier of the 1D migration is much smaller than the rotation barrier. Therefore, trapping and de-trapping (DT process) of SIA clusters would result in confinement of their 1D-glide between traps [17,18]. On the other hand, impurities or solutes can reduce the rotation barrier, [19,20], allowing SIA clusters to effectively diffuse in 3D (net-3D diffusion). In this paper, we compare the 1D and net-3D cases in terms of effectiveness in facilitating SIA-vacancy recombination.

To investigate the effect of the above two cases of SIA diffusion, we performed object Kinetic Monte Carlo (OKMC) simulations of damage accumulation in polycrystalline tungsten under 14 MeV neutron irradiation. The simulations were carried out using the KSOME (kinetic simulation of microstructure evolution) code [21, 22]. Cascade debris from molecular dynamics simulations with primary knock-on atom (PKA) energies corresponding to 14 MeV neutrons was taken as the source of primary defects for the OKMC simulations. The damage accumulation was simulated at 1025 K and as a function of dose rate, de-trapping barrier, and impurity concentration. In the net-3D case, the glide direction after de-trapping is random for all sizes of SIA clusters. In the confined-1D case, the glide direction after de-trapping is random only for SIA clusters with size \leq 5, while SIA clusters larger than size 5 retain their original glide direction. In both cases of simulations, trapped vacancies are assumed to be stable and permanently immobilized. In both cases of simulations, trapped vacancies are assumed to be stable and permanently immobilized, but the self-trapped vacancy clusters were allowed to dissolve via mono-vacancy emission

Unexpectedly, the damage accumulation was lower with confined-1D diffusion than that with net-3D at the lowest dose (10^{-8} dpa /s) rate and the trap concentration (100 appm) studied. However, for the same trap concentration, at the highest dose rate studied (10^{-4} dpa/s) , the opposite was true. For both cases of SIA diffusion, the damage accumulation first increases from 100 to 150 appm, but then the damage either decreases or saturates from 150 to 200 to 250 appm. Furthermore, the difference in the damage accumulation between the two cases diminishes with increasing trap concentration.

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9.2 OKMC SIMULATIONS OF He IRRADIATION OF TUNGSTEN SURFACE: CALCULATION OF DEPTH DEPENDENT ACTIVATION ENERGY BARRIERS OF TRAP MUTATION REACTIONS IN THE NEAR-SURFACE REGION OF W(100) SURFACE—Giridhar Nandipati, Kenneth J. Roche, Richard J. Kurtz, Wahyu Setyawan (Pacific Northwest National Laboratory), Karl D. Hammond (University of Missouri), Dimitrios Maroudas (University of Massachusetts), Brian D. Wirth (University of Tennessee)

OBJECTIVE

This task aims to use object kinetic Monte Carlo (OKMC) simulations to study the evolution of the heliumbubble microstructure in the near-surface region of plasma-exposed tungsten surface. Kinetic simulations of microstructural evolution (KSOME) [1,2], an OKMC code is used to simulate spatially relevant nearsurface microstructures to experimentally relevant time-scales under isothermal and variable temperature conditions caused by Edge-Localized Modes (ELMs).

SUMMARY

This report documents the calculation of depth-dependent activation energy barriers and prefactors of trapmutation processes using their relative occurrence probabilities reported in Refs. 3 & 4 for He clusters of size 2 to 7 in the near-surface region of W(100) surface.

PROGRESS AND STATUS

Introduction

In the studies reported in References 3 & 4, several hundred MD simulations of the dynamics of isolated He_n clusters (n =size) towards the free surface were performed. Individual outcomes of these simulations are the trap mutation (TM) or TM followed by partial dissociation (PD) reactions undergone by the clusters at various depths or the escape of the entire cluster to the gas phase (D). The outcome of individual MD simulation runs for the He_n ($2 \le n \le 7$) clusters were recorded to obtain the overall occurrence probabilities of individual reactions. Accordingly, for a given Hen cluster size, occurrence probabilities for TM events at a particular depth reported in References 3 & 4 depends on the occurrence probabilities of those that occur at lower depths. However, to calculate the activation energy barriers and prefactors for these reactions at a particular depth, probabilities of all competing processes (referred to as absolute probabilities) at that depth are required. Accordingly, the sum of these absolute probabilities



Figure 1. P_i^r , where $i = 1, \dots, 8$, are the relative occurence probabilities of TM events that occur at various depths. P_5^a is the absolute occurrence probability for a TM event that occurs at the depth of I = 5. P_5^d is the absolute probability for the diffusion of the He cluster from the depth of I = 5 to I = 4.

should be equal to one. Absolute occurrence probabilities for TM events at a given depth can be calculated as shown in Figure 1. More importantly, as the diffusion of a He_n cluster towards the surface is the main competing process with TM events, the probability of non-occurrence of TM events is taken as the occurrence probability for the He_n cluster to hop to an upper layer. Furthermore, for all cluster sizes, the

probability of complete desorption of a He_n cluster $W + He_n \rightarrow W + nHe_{(g)}$ is taken as the probability for the He_n cluster to diffuse to the upper layer. Note that when the He_n cluster is at a depth of *I*=3, because of the elastic interaction with the free surface, its activation barrier is 0 eV and its rate equal to the diffusion prefactor for that particular He cluster size.

Expressions Used Calculate Activation Barrier and Prefactors

When the average time scale (Δt_{avg}) or the average rate (R_T) of a process at a given temperature T and its activation barrier (E_a) are known, then the prefactor (D_0) is expressed using Equation (1)

$$D_0 = \frac{1}{\Delta t_{avg}} \exp\left(\frac{E_a}{k_B T}\right) = R_T \exp\left(\frac{E_a}{k_B T}\right) \qquad (1)$$

However, if the rate R_T of a process at a given temperature T and its prefactor are known then the activation barrier E_a for this process can be calculated using Equation (2)

$$E_a = k_B T \ln\left(\frac{D_0}{R_T}\right) \tag{2}$$

If the rates of a process R_{T_1} and R_{T_2} at two different temperatures T_1 and T_2 are known, then the activation barrier for this process can be calculated using Equation (3)

$$E_{a} = \frac{k_{B}T_{2}T_{1}}{T_{2} - T_{1}} \ln\left(\frac{R_{2}}{R_{1}}\right)$$
(3)

If the ratio of the rates $(\mathcal{R}_{\mathcal{T}}^{ab})$ of two competing processes L_a and L_b at two different temperatures T_1 and T_2 are known, then the difference of their activation energy barriers is given by Equation (4)

$$\Delta E_{ba} = E_b - E_a = \frac{k_B T_1 T_2}{T_2 - T_1} \ln\left(\frac{\mathcal{R}_{T_1}^{ab}}{\mathcal{R}_{T_2}^{ab}}\right)$$
(4)

where $\mathcal{R}_{T_i}^{ab} = \frac{R_a}{R_b}\Big|_{T_i}$ at a temperature T_i where *i*=1,2.

Activation Energy Barriers and Prefactors for Trap Mutation Reactions

He₂ Cluster

Table 1. List of reactions undergone by the He2 cluster (of size 2) near the W(100) surface and their relative probabilities (*I=1* corresponds to the surface layer) taken from Reference 2

ID	$W + He_2 \rightarrow$	1000 K	1200 K
L ₁	$He_2V(I = 4) + W_s$	0.6%	0.8%
L ₂	$He_2V(l = 3) + W_s$	74.3%	74.2%
L ₃	HeV $(I = 3) + W_s + He_{(g)}$	4.8%	5.9%
L4	(/=3) W+2He _(g)	20.3%	19.1%
Num. Traj.		522	288

Process L₁

For the TM reaction L_1 , the competing process is the diffusion of He_2 cluster from l=4 to l=3, i.e., towards the surface. Hence, the occurrence probabilities for the diffusion hop at 1000 and 1200 K are 0.994 and 0.992, respectively. The activation energy barriers for He_2 cluster diffusion in the bulk at temperatures 0, 1000 and 1200 K are 0.26, 0.401, and 0.454 eV, respectively. [5]. However, taking into account the elastic interaction of the He_2 cluster with the free surface, [6,7] the activation energy barriers for this cluster's diffusion from l=4 to l=1 at 1000 and 1200 K are 0.323 eV and 0.365 eV, respectively. Since the ratio of

rates of two competing processes at two different temperatures is known, the activation barrier and the prefactor for reaction L_1 can be calculated using Equation (4) to be 0.48 eV and 4.1×10^{10} /s, respectively. Note that, for simplicity, the diffusion to the layer l=4 and away from the surface are ignored. Furthermore, the difference in the activation barrier for the diffusion hops at 1000 and 1200 K is also ignored, as it is only 0.04 eV.

Processes L_2 , L_3 and L_4

Since L_2 , L_3 , and L_4 occur at the same depth, they can be considered as competing processes, with L_3 being the diffusion process. From Figure 1 in Reference [3], Δt_{avg} for L_2 is taken as 25 ps, which corresponds to a rate of 4×10^{12} /s at 1000 K, and its activation barrier is 0.31 eV. Following the same approach used for the process L_1 , the activation energy barriers and prefactors for L_3 and L_4 can be calculated. A complete list of the calculated activation energy barriers and prefactors is shown in Table 3.

 L_3 and L_4 at I=3

ID	$\textbf{W+He}_2 \rightarrow$	1000 K	1200 K
L ₂	$He_2V (I = 3) + W_s$	74.75%	74.8%
L ₃	HeV ($I = 3$) + W _s +He _(g)	4.84%	5.95%
L_4	HeV ($I = 3$) + W+2He _(g)	20.42%	19.25%

Table 2. Absolute probabilities for the processes L₂, Table 3. Calculated activation energy barriers and prefactors of the TM reaction events of He₂

ID	W + He ₂ \rightarrow	D ₀ (s ⁻¹)	E _a (eV)
L ₁	$He_2V (I = 4) + W_s$	4.1×10^{10}	0.48
L ₂	He_2V (/ = 3) + W_s	1.45 × 10 ¹²	0.31 [<mark>1</mark>]
L3	$HeV (I = 3) + W_s + He_{(g)}$	3.4 × 10 ¹¹	0.42
L4	(<i>I</i> = 3) W+2He _(g)	8.143 × 10 ¹⁰	0.28

He3 He4 He5, He6 and He7 Clusters

Activation energy barriers and prefactors of TM reactions for the remaining cluster sizes were calculated following the same approach.

Table 4. Activation barriers and prefactors for the TM reactions of the He_3 cluster

ID	$\text{He}_3 \rightarrow$	$D_0(s^{-1})$	E_a (eV)
L_1	$He_3V(l=5)$	5.313×10^{11}	0.532
L_2	$He_3V(l=4)$	9.38×10^{14}	0.5 [3]
L_3	$He_2V(l=3) + He_{(g)}$	1.13×10^{16}	0.783
L_4	$HeV(l = 3) + 2He_{(g)}$	1.31×10^{15}	0.378
L_5	$3He_{(g)}$	3.73×10^{12}	0.00

Table 6. Activation barriers and prefactors for the TM reactions for the He_5 cluster

ID	W+He₅ →	Ea(eV)	D₀(s ⁻¹)
L_3	$He_5V_2(l=4)$	0.49	10 ¹³
L_4	$He_3V_2(l=4) + 2He_{(g)}$	0.22	10 ¹³
L_5	$He_2V(l=4) + 3He_{(g)}$	0.25	10 ¹³
L_6	$HeV(l = 4) + 4He_{(g)}$	0.3	10 ¹³
7	F 11 -	0 0	1 2 0 1011

Table 8. Activation barriers and prefactors for the TM reactions for the He_7 cluster

ID	He ₇ →	E(eV)	D₀(s⁻¹)
L_2	$He_7V_2(l=5)$	0.28 [3]	3.75×10^{12}

Table 5. Activation barriers and prefactors for the TM reactions of the He_{1} cluster

ID	$\text{He}_4 \rightarrow$	E_a (eV)	D ₀ (s ⁻¹)
L_2	$He_4V \ (l=5)$	0.48	10 ¹²
L_3	$He_4V \ (l=4)$	0.21 [3]	2.92×10^{13}
L_4	$He_2V(l=3) + 2He_{(g)}$	-	-
L_5	$HeV (l = 3) + 3He_{(g)}$	0.084	1014
L_6	$4\text{He}_{(g)}$	0	1.9×10^{13}

Table 7. Activation barriers and prefactors for the TM reactions of the He₆ cluster

ID	W+He₅ →	Ea(eV)	D ₀ (s ⁻¹)
L_2	$He_6V(l=6)$	0.89	1012
L_3	$He_6V_2(l=5)$	0.40	10 ¹²
L_4	$He_6V_3(l=4)$	0.09	10 ¹³
L_5	$He_3V_2(l=4) + 3He_{(g)}$	0.05	10 ¹³
L_6	$He_2V(l=4) + 4He_{(g)}$	0.11	10 ¹³
L_7	$HeV(l = 4) + 5He_{(g)}$	0.37	10 ¹³
L_8	$6He_{(g)}$	0.0	1.53×10^{13}

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9.3 A COMPUTATIONAL METHOD USING THREE-DIMENSIONAL FINITE ELEMENT ANALYSES TO PREDICT BRICK-AND-MORTAR'S ELASTIC PROPERTIES—Ba Nghiep Nguyen, Cleopatra A Howell, Charles H. Henager Jr., Wahyu Setyawan (Pacific Northwest National Laboratory)

OBJECTIVE

The objective is to investigate and model the mechanical properties of ductile phase toughened tungsten (W) composites that possess hierarchical "brick-and-mortar" (BAM) microstructures. During the current reporting period, a rigorous computational method based on three-dimensional (3D) finite element (FE) analyses was developed to accurately and completely predict all the elastic properties of a BAM material.

SUMMARY

A computational method was developed to compute each unknown component of the 6x6 stiffness tensor of a composite microstructure through FE analyses with associated boundary conditions. This method was then applied to a BAM 3D microstructure having two hierarchical levels: Level 0 corresponds to a single W "brick" with length L and height H; Level 1 includes the representative W-brick-and-Ni/Fe "mortar" unit cell that can be repeated in three directions of a structural coordinate system to build the microstructural BAM domain. The determination of the stiffness tensor allows us to obtain all the elastic properties of a BAM composite.

PROGRESS AND STATUS

Introduction

Biological materials such as nacre, teeth, and shells have been found to exhibit attractive and unique combinations of stiffness, strength, and fracture toughness. Research has already been conducted in various engineering areas to mimic the kind of naturally occurring hierarchical microstructures or nanostructures of biological materials to produce materials with optimum stiffness, high strength and fracture toughness for their intended structural applications. In the same objective, we have investigated the mechanical properties of W composites that possess hierarchical BAM microstructures.

The W-allovs and W-composites are being studied as potential materials for plasma-facing components (PFCs) of future fusion reactors due to their high melting point, strength at high temperatures, high thermal conductivity, low coefficient of thermal expansion, and low sputtering yield [1-4]. However, W and most Walloys exhibit rather low fracture toughness and a high DBTT that would render them as brittle materials during reactor operations [2,5]. The DBTT for unirradiated W-alloys typically ranges from 573K to 1273K (300°C to 1000°C), and in a reactor environment radiation hardening would further elevate this range [4,6-7]. W-alloys toughened by engineered reinforcement architectures are therefore needed and are strong candidates for PFCs. Recently, there is a strong interest in W composites having hierarchical microstructures such as W BAM composites as these materials can exhibit optimum combinations of stiffness, strength and fracture toughness required for applications in PFCs. In the previous guarters, we had investigated the mechanical properties of BAM composites in terms of selected relevant microstructural parameters, i.e., microstructural topology, brick aspect ratio, brick distribution over a prescribed domain. Linear and nonlinear two-dimensional FE analyses were performed to study the effects of these parameters on the composite stress/strain response and strength [8-10]. During this reporting period, a rigorous computational method based on 3D FE analyses was developed to accurately and completely predict all the elastic properties of any BAM material as these properties are needed and required in any linear and nonlinear structural analyses using BAM composites at the macroscopic scale of a PFC.
Model Development

Figure 1a shows the 3D FE mesh for a BAM unit cell microstructure. The volume domain of this unit cell with the definition of the surfaces for prescribed boundary conditions (BCs) is given in Figure 1b. The problem consists of finding in the domain volume the stress (σ) and displacement (\mathbf{u}) fields that satisfy:

Equilibrium:
$$div \sigma = 0$$
 (1)
Constitutive relation: $\sigma = \mathbf{C} : \boldsymbol{\varepsilon}(\mathbf{u})$ (2)

where ε is the strain tensor caused by the displacement field, **u**; and all the bold notations denote tensor quantities. The 3D FE method is used to solve systems (1) and (2) with the associated BCs allowing to extract each component of the fourth-order *homogenized* elastic stiffness tensor, **C** of the BAM unit cell. Therefore, a priori the number of FE analyses that needs to be performed exactly corresponds to the number of nonzero components of **C**. However, exploring the *symmetry* of the stiffness tensor and the *orthotropic symmetry* assumed for the BAM microstructure, there are only nine unknown components of **C** that need to be obtained by the analyses to completely determine this tensor. These nonzero unknown components are: $C_{11}, C_{22}, C_{33}, C_{12}, C_{13}, C_{23}, C_{44}, C_{55}, and C_{66}$ written using contracted indices. Once the stiffness **C** has been determined, the inverse of **C** is the compliance tensor **S** from which all the elastic properties are directly obtained in terms of the components of **S**.



Figure 1. (a) A BAM unit cell discretized in 3D FE elements (brick regions in gray and mortar region in green) and (b) the corresponding unit cell domain with defined surfaces for prescribed boundary conditions (BCs).

Computation of C11

The following BCs are prescribed to the unit cell domain (Figure 1b) to extract C_{11} through an elastic FE analysis:

- On S_{+x}: Applying uniform displacements along x, Δx ,
- On S-x: Applying uniform displacements along x, $-\Delta x$,
- On S+y: Fixing displacements along y,
- On S-y: Fixing displacements along y,
- On S+z: Fixing displacements along z,
- On S_{-z}: Fixing displacements along z.

These BCs yield:

$$C_{11} = \frac{\langle \sigma_{xx} \rangle}{\langle \varepsilon_{xx} \rangle} \tag{3}$$

where $\langle \sigma_{xx} \rangle$ and $\langle \varepsilon_{xx} \rangle$ are the average σ_{xx} stress and ε_{xx} strain over all the elements of the volume domain. σ_{xx} and ε_{xx} are computed by the FE analysis. The Δx and $-\Delta x$ are constant displacement values of the same magnitude but opposite sign applied on S_{+x} and S_{-x} in the x-direction, respectively. In the remainder of this report, any variable between brackets '<.>' denote its corresponding average value in the domain.

Computation of C22

In a similar manner, the following BCs are prescribed to the unit cell domain to obtain C_{22} :

- On S+x: Fixing displacements along x,
- On S-x: Fixing displacements along x,
- On S_{+y}: Applying uniform displacements along y, Δy ,
- On S_{-y}: Applying uniform displacements along y, $-\Delta y$,
- On S+z: Fixing displacements along z,
- On S_{-z}: Fixing displacements along z.

These BCs yield:

$$C_{22} = \frac{\langle \sigma_{yy} \rangle}{\langle \varepsilon_{yy} \rangle} \tag{4}$$

Computation of C₃₃

The following BCs are prescribed to the unit cell domain to obtain C_{22} :

- On S_{+x}: Fixing displacements along x,
- On S-x: Fixing displacements along x,
- On S+y: Fixing displacements along y,
- On S_{-y}: Fixing displacements along y,
- On S_{+z}: Applying uniform displacements along z, Δz ,
- On S_{-z}: Applying uniform displacements along z, -Δz.

These BCs yield:

$$C_{33} = \frac{\langle \sigma_{zz} \rangle}{\langle \varepsilon_{zz} \rangle} \tag{5}$$

Computation of C12

The prior computation of C_{11} (see above) allows the determination of C_{12} in a subsequent FE analysis using the following BCs prescribed to the unit cell domain:

- On S_{+x}: Applying uniform displacements along x, Δx ,
- On S_{-x}: Applying uniform displacements along x, $-\Delta x$,
- On S+y: Free,
- On S-y: Free,
- On S+z: Fixing displacements along z,
- On S-z: Fixing displacements along z

These BCs yield:

$$C_{12} = \frac{\langle \sigma_{xx} \rangle - C_{11} \langle \varepsilon_{xx} \rangle}{\langle \varepsilon_{yy} \rangle}$$
(6)

Computation of C₁₃

Similarly, a subsequent FE analysis using the following BCs prescribed to the unit cell domain allows to obtain C_{13} :

- On S_{+x}: Applying uniform displacements along x, Δx,
- On S_{-x}: Applying uniform displacements along x, $-\Delta x$,
- On S_{+y}: Fixing displacements along y,
- On S_{-y}: Fixing displacements along y,
- On S_{+z}: Free,
- On S-z: Free.

These BCs yield:

$$C_{13} = \frac{\langle \sigma_{xx} \rangle - C_{11} \langle \varepsilon_{xx} \rangle}{\langle \varepsilon_{zz} \rangle}$$
(7)

Computation of C23

The prior computation of C_{22} (see above) allows the determination of C_{23} in a subsequent FE analysis using the following BCs prescribed to the unit cell domain:

- On S+x: Fixing displacements along x,
- On S-x: Fixing displacements along x,
- On S_{+y}: Applying uniform displacements along y, Δy
- On S_{-y}: Applying uniform displacements along y, $-\Delta y$
- On S+z: Free,
- On S-z: Free.

These BCs yield:

$$C_{23} = \frac{\langle \sigma_{yy} \rangle - C_{22} \langle \varepsilon_{yy} \rangle}{\langle \varepsilon_{zz} \rangle}$$
(8)

Computation of C₄₄

 C_{44} corresponds to the shear modulus G_{yz} . It can be determined using the following BCs:

- On S+x: Free,
- On S-x: Free,
- On S+y: Fixing displacements along z,
- On S-y: Fixing displacements along z,
- On S_{+z} : Applying uniform displacements along y, Δy ,
- On S-z: Applying uniform displacements along y, $-\Delta y$.

These BCs cause shear stresses τ_{yz} and shear strains γ_{yz} in the unit cell domain allowing to obtain:

$$C_{44} = G_{yz} = \frac{\langle \tau_{yz} \rangle}{\langle \gamma_{yz} \rangle} \tag{9}$$

Computation of C₅₅

 C_{55} corresponds to the shear modulus G_{xz} . It can be determined using the following BCs:

- On S+x: Fixing displacements along z,
- On S-x: Fixing displacements along z,
- On S+y: Free,
- On S_{-y}: Free,
- On S+z: Applying uniform displacements along x, Δx,
- On S_{-z}: Applying uniform displacements along x, $-\Delta x$.

These BCs yield:

$$C_{55} = G_{xz} = \frac{<\tau_{xz}>}{<\gamma_{xz}>}$$
(10)

Computation of C₆₆

 C_{66} corresponds to the shear modulus G_{xv} . It can be determined using the following BCs:

- On S+x: Fixing displacements along y,
- On S-x: Fixing displacements along y,
- On S_{+y}: Applying uniform displacements along x, Δx ,
- On S_{-y}: Applying uniform displacements along x, $-\Delta x$,
- On S_{+z}: Free,
- On S-z: Free.

These BCs yield:

$$C_{66} = G_{xy} = \frac{<\tau_{xy}>}{<\gamma_{xy}>}$$
(11)

Results

This section illustrates the application of this method to determine the elastic properties of the BAM material (illustrated in Figure 1a) made of W bricks and the Ni-Fe mortar. The elastic modulus and Poisson's ratio of W used in the analyses are 383,000 MPa and 0.28 while those for Ni-Fe are 203,570 MPa and 0.304, respectively. Nine 3D FE analyses were conducted using the FE mesh given in Figure 1a with the associated BCs discussed above to determine the nine unknown components of the composite stiffness tensor. They are reported below in MPa:

 $C_{11} = 441212; C_{22} = 427677; C_{33} = 444500$ $C_{12} = 169857; C_{13} = 172942; C_{23} = 170462$ $C_{44} = 127839; C_{55} = 133763; C_{66} = 126934$

The results show that $C_{11} \approx C_{33}$ and $C_{44} \approx C_{66}$. These confirm the orthotropy assumption for this BAM composite, and the material orthotropy is nearly realized about the y-axis of the coordinate system shown

in Figure 1. Next, the compliance tensor is obtained by taking the inverse of the stiffness tensor allowing to obtain all the elastic properties in terms of the compliance tensor components. The results for elastic properties are:

Elastic moduli (MPa): E_{xx} = 344338; E_{yy} = 333639; E_{zz} = 346924 Shear moduli (MPa): G_{yz} = 127839; G_{xz} = 133763; G_{xy} = 126934 Poisson's ratios: v_{xy} = 0.2858; v_{xz} = 0.2795; v_{zy} = 0.2867

Conclusions

During this reporting period, a rigorous computational method based on 3D FE analyses was developed to accurately and completely predict all the elastic properties of a BAM material. The developed method is general and can be applied to compute the elastic properties of any 3D W-based composite having any as-formed microstructure. In the case of regular lattice BAM microstructures, the BAMs are generally nearly orthotropic, and assuming such an orthotropy allows to reduce the number of FE analyses to 9. This work is a significant step towards designing BAM composites for fusion materials applications as the elastic properties are always needed and required in any linear and nonlinear structural analyses using W BAM composites or in general W-based composites at the macroscopic scale of a PFC.

Future Work

A journal paper entitled: "*Tailoring Ductile-Phase Toughened Tungsten Hierarchical Microstructures for Plasma-Facing Materials*" by B.N. Nguyen and C.H. Henager Jr. is being completed to describe our previous nonlinear analyses of W BAM composites [10]. We will also write a journal paper reporting this computational method for elastic properties of BAM materials. Further elastic-plastic damage analyses will be conducted upon availability of experimental data for W/Ni-Fe tensile specimens.

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9.4 DENSITY FUNCTIONAL THEORY STUDY OF HELIUM AND HYDROGEN DISTRIBUTION IN W-Ni-Fe HEAVY ALLOYS—W. Setyawan (Pacific Northwest National Laboratory)

OBJECTIVE

The overall objective is to provide scientific understanding of the effect of fusion neutron irradiation, helium, and hydrogen isotopes on the thermo-mechanical properties of ductile-phase-toughened (DPT) W-Ni-Fe heavy alloys, as well as understanding of He and H gas retention in these DPT alloys. The specific objective in this report is to determine whether He will segregate to the W/Ni-Fe-W interphase boundary.

SUMMARY

Density functional theory (DFT) method is employed to calculate the formation energies of a He atom and a H atom at various interstitial sites in an interphase boundary (IB) between bcc W and fcc Ni. The fcc Ni is a surrogate for the Ni-Fe-W solid solution (the ductile phase) of a W-Ni-Fe ductile-phase toughened W composite. An interphase boundary model with an orientation relationship of {110}<112>Ni//{110}<111>W is employed for this study based on an experimental observation. The DFT data show that He is more stable at the IB than in Ni (and least stable in W), while H is more or less as stable in the IB as in Ni (and least stable in W). Future research includes exploring the effect of Fe and W solute in Ni particularly at the IB to simulate the solid solution.

PROGRESS AND STATUS

Introduction

The DPT W composites derived from W-Ni-Fe heavy alloys (WHAs) are being researched as potential plasma-facing materials (PFMs). The alloys typically consist of essentially pure body-centered cubic (bcc) W particles embedded in ductile face-centered cubic Ni-Fe-W solid solution matrix (binder) phase [1-2]. The WHAs are cheaper than W and commercially available. Cohesion of the interphase boundary between the particle and the binder is key to the excellent ductile-phase toughening in these materials. The excellent toughness of WNiFe DPT composites has been demonstrated, for instance, from tensile tests of 95W-3.5Ni-1.5Fe alloy (wt. composition) alloy (referred to as 95W) [3] and 97W-2.1Ni-0.9Fe alloy (97W) [4] showing that these composites exhibit an ultimate strain of ~12% at room temperature. Further evidence of the toughening is demonstrated in bend tests of 90W-7Ni-3Fe alloy (90W) [1] which show a peak stress intensity of ~52 MPa/ \sqrt{m} at room temperature (with a displacement rate of 0.2 mm/min), significantly higher than 8 MPa/ \sqrt{m} of a monolithic W.

For use as PFMs, the effects of neutron irradiation, helium, and hydrogen isotopes need to be understood. For instance, whether or not He and H will segregate to the interphase boundaries, and if they do, how the interphase cohesion is affected. Furthermore, the effect of atomic displacement damage from neutron irradiation on the phase stability of the solid solution against precipitation of brittle intermetallic compounds and the stability of the interphase boundaries need to be explored. Neutron activation of Ni also needs to be quantified to determine the upper limit of Ni content allowed in the alloys. In regard to the neutron activation of Ni, a study has been performed to assess the activation of the 97W as a first wall material in a DEMO concept reactor [2]. The study shows that since 97W contains only a small amount of Ni (2 wt.%), the dose rate of the irradiated 97W after 1 year of operation is very similar to that of pure W, and it is less than two times higher than W after 10,000 years. The analysis suggests that 97W alloys are acceptable for fusion application.

Results

In this report, DFT method is employed to determine whether He will segregate at the interphase boundaries between the W particles and the binder phase. Data obtained from electron diffraction spectroscopy (EDS) show that the composition of the binder among the different alloys is similar, namely approximately 66 at.%

Ni, 25 at% Fe, and 9 at% W. To understand the general preferential distribution of He in the DPT system, pure Ni is taken in place of Ni-Fe-W for the binder phase. Hence, the interphase boundary (IB) is modeled between bcc W and fcc Ni. The orientation relationship (OR) of the IB model is taken based on the transmission electron spectroscopy study by Zhu et al. [5] who proposed an incoherent interphase boundary with an OR of {-220}<112>binder//{-220}<111>W.

The DFT calculations are performed with VASP.5.4.1 software. Generalized gradient approximation (GGA) with Perdew-Burke-Ernzerhof (PBE) functionals are employed for the electronic exchange and correlation energies. Projector augmented wave (PAW) pseudopotential formalism is employed to describe the ionelectron interactions. The pseudopotentials for Ni, W, and He are taken from VASP library potpaw_PBE.54. The number of electrons treated as valence is 10 (Ni), 6 (W), and 2 (He). Plane wave energy cutoff of 620 eV is employed. A Monkhorst-Pack 3x3x1 **k**-point mesh is employed to sample the Brillouin zone. Simulation cells and atomic positions are fully relaxed. Structure relaxation is performed with a total energy convergence of 5 meV. In this early study, electronic spin is not taken into account in all simulations.

Interphase Boundary Model

Based on the OR of the IB as mentioned above, a model is constructed with 6 layers of W{110} and 9 layers of Ni{110} with W[111]//Ni[112] and W[-1 -1 2]//Ni[-1 -1 1]. The simulation cell is oriented such as the W[111] is along the x-axis, W[-1 -1 2] is along the y-axis, and the slab normal is along the z-axis. First, based on the equilibrium lattice constant of W, a 6-layer W{110} slab is created with a surface dimension of 8.241x7.770 Å² (i.e. 8.241 Å along x and 7.770 Å along y). Likewise, a 9-layer Ni{110} slab is created with a surface dimension of 8.614x6.091 Å². To construct the initial IB model, the dimension of the Ni slab is rescaled to that of the W slab and placed on top of the W slab. A vacuum of ~16 Å is added in the simulation cell to eliminate the interaction between the top and bottom free surfaces across the periodic z-boundary. A set of initial structures is generated by shifting the Ni slab with respect to the W slab along the surface dimensions (8 grids along each x and y dimensions). The initial structures are then fully relaxed. Figure 1 shows the structure of the most stable configuration which is then taken as the IB model subsequent calculations. The relaxed surface dimension is 8.300x7.347 Å².



Figure 1. Structure of the most stable configuration of $\{110\}<112>Ni/{\{110}<111>W$ slab that is taken as the IB model in this study. The slab is oriented with W[111]//Ni[112] along x and W[-1 -1 2]//Ni[-1 -1 1] along y. a) view from the y direction, b) view from the x direction, c) view from the z direction. There are 9 W atoms per W{110} layer and 6 Ni atoms per Ni{110} layer. In c), only the W and Ni layers closest to the interphase boundary are shown. Bonds < 3 Å are drawn.

Interstices within the interphase boundary region are then searched using the particle-swarming method. The region is divided into 11x11x6 grids and the center of each grid is taken as the location of the initial swarming particles (i.e. 726 initial particles). For each particle, the four nearest atoms (Ni and/or W) are determined and the geometrical center of these atoms is taken as the position of the particle. Subsequently, redundant particles are removed based on a cutoff radius of 1 Å. There are 24 unique interstices (sites) found and shown in Figure 2.



Figure 2. Location of interstices within the IB model as labelled 1 to 24 used to study He and H segregation. Bonds between each interstice to the Ni and W atoms within 2 Å are drawn.

The segregation of He towards the IB is studied by placing a He atom in one of the sites. The formation energy (E_f) of a He atom at site-*i* is then calculated as the following

$$E_{f}$$
{He,IB,i} = E {He,IB,i} - E {IB} - E {He}} (1)

Where E{He,IB,i} is the total energy of the IB system with He at site-*i*, E{IB} is the total energy of the pure IB system (i.e. without a He atom), and E{He} is the total energy of an isolated He atom. To calculate the segregation energy, the formation energy of He in bulk W and bulk Ni needs to be determined. Using a similar simulate cell as the IB system but containing only W{110} slab with 10 layers, He is found to be most stable at a tetrahedral site with a formation energy of E_{f} {He,W,tet} = 6.39 eV. Similarly, using only Ni{110} slab with 13 layers, He is also found to be most stable at a tetrahedral site with a formation energy (E_{seg}) of a He atom at site-*i* in the IB is then calculated with respect to the tetrahedral site in bulk Ni as the following

$$Eseg\{He, IB, i\} = E_{f}\{He, IB, i\} - Ef\{He, Ni, tet\}$$
(2)

Figure 3a shows the segregation energy of He with respect to bulk Ni as a function of the IB site. In all cases, the segregation energy is negative, indicating that He is more stable at the IB than in bulk Ni. On the other hand, He in W is less stable than in Ni. The preference distribution of H is also studied. For H, the formation energy is calculated using a similar formula as Eq 1, however the total energy of H-only system is taken as a half of the total energy of a diatomic H₂ molecule instead of an isolated H atom. In W, like He, H is most stable at the tetrahedral site (E_{f} (H,W,tet) = 0.95 eV). However, in Ni, H is most stable at the octahedral sites (E_{f} (H,Ni,oct) = -0.02 eV). The segregation energy of H is calculated with respect to H at the octahedral site in Ni. The results are shown in Figure 3b. The segregation energy of H is negative in some cases but positive in other cases, indicating that H is more or less as stable at the IB as in Ni. On the other hand, H in W is less stable than in Ni or in the IB.



Figure 3. Segregation energy (E_{seg}) of a) He and b) H with respect to bulk Ni as a function of the IB site. Negative values indicate tendency to segregate towards the IB. The reference site in Ni is the tetrahedral site for He and the octahedral site for H.

Future Research

Future research includes 1) taking into account the effect of electronic spin, 2), exploring the effect of Fe and W solute at the interphase boundary, 3) exploring the effect of vacancy and interstitial, 4) exploring the change in the cohesion energy of the interphase boundary due to He and H, 5) exploring different interphase boundaries, and 6) comparison with He ion-beam irradiation experiments.

Acknowledgement

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10. **FUSION SYSTEM DESIGN**

No contributions this reporting period.

11. **IRRADIATION & TESTING METHODS, EXPERIMENTS AND SCHEDULES**

11.1 IRRADIATION CREEP OF F82H PRESSURIZED TUBES—A.A. Campbell, Y. Katoh, J.W. Geringer, J.D. Reed, K.R. Smith, P.L. Mulligan (Oak Ridge National Laboratory), M. Ando (QST)

OBJECTIVE

The objective of this task was the measurement of neutron irradiation-induced creep of reduced activation ferritic martensitic (RAFM) steel alloy F82H at 300°C. The stress necessary to drive irradiation creep was provided using thin-walled pressurized tubes. The internal inert gas pressure induces a hoop stress in the sealed tubes. The hoop stress drives irradiation-induced creep in the tube, which is measured as changes in the tube diameter.

SUMMARY

Four creep capsules designed for a target irradiation temperature of 300°C and hoop stresses of 0, 150, 300, and 380 MPa [1], were irradiated in the HFIR during cycles 475 and 476. Disassembly and postirradiation dimensional inspection of the four creep tube samples was completed.

PROGRESS AND STATUS

The neutron irradiation creep of RAFM steels, such as F82H, will be important for the design of fusion reactor blanket systems. This work is redeveloping the capability at ORNL to perform pressurized creep tube (PCT) experiments in the flux trap of HFIR to accelerate neutron damage and the capability to measure the creep deformation.

The four capsules completed two irradiation cycles in HFIR in December 2017, with a nominal radiation damage of 3.7 dpa. The capsules were shipped from the HFIR to the Irradiated Materials Examination and Testing (IMET) hot cells facility. The capsule disassembly and post-irradiation exanimation (PIE) work was performed in the hot cells in the IMET facility.

The capsule disassembly and PIE procedure followed the following steps:

- 1. Puncture outer capsule housing determine whether pressurization gas inside the PCTs was retained.
- 2. Cut open capsule and extract specimen and holder thermometry.
 - a. Holder thermometry put aside for shipment to Low Activation Materials Development and Analysis Laboratory (LADMA) for analysis.
- 3. Microscope investigation of PCT outer surfaces, ensure no large visible flaws.
- 4. Measuring outer diameter of PCTs while pressurized, via 2 non-contact techniques.
 - a. Laser profilometry using a Beta LaserMike model 162.
 - b. Optical imaging with a Celestron hand-held microscope.
- 5. Cut ends off specimen to release pressurizing gas and extract internal sample thermometry. a. Internal sample thermometry put aside for shipment to LADMA for analysis.
- 6. Remeasure outer diameter of PCTs via same methods in Step 4.

Two pieces of equipment were designed for this work, the capsule puncture system (Step 1) and a stage for the optical imaging with the hand-held microscope (Step 4b). The capsule puncture system (Figure 1a) is a self-contained pressure vessel that has a puncture tip that puts a hole in the capsule outer aluminum housing and a pressure gauge for measuring if any gas escapes from the capsule. The composite imaging with the microscope (Figure 2b) has an articulating stage that moves the sample a known distance between images, and includes a gauge pin and millimeter gauge for calibration.



Figure 1. (a) system for puncturing capsule housing. (b) setup for composite optical imaging with a microscope.

The capsule puncture step found that the specimens pressurized at 300 MPa and 380 MPa did not retain their pressurization gas, while the 0 MPa and 150 MPa specimens were still intact. Capsule disassembly for the intact PCTs only required a shake of the capsule to remove the specimens, while the failed PCTs required that the inner V-4Cr-4Ti holder be milled open for PCT extraction. The corrugated aluminum foil between the PCT and the holder (to promote even heat transfer) was easily removed from the intact PCTs, while the failed specimens required extra work to remove the foil. After capsule disassembly the failed specimens were investigated, which found a rupture in the thin-wall region of the 380 MPa PCT (Figure 2), while no such failure indication was observed in the 300 MPa PCT.



Figure 2. Image of sample pressurized at 380 MPa showing rupture location.

The failure of the 300 MPa and 380 MPa specimens is still unknown. It is possible that the specimens failed early in irradiation because at 573 K the pre-irradiation yield strength is ~480 MPa so the 300 MPa and 380 MPa internal stresses were a high percentage of the yield strength. If the irradiation temperature was higher than 573 K the pre-irradiation yield stress would have been reduced, making the possibility of failure higher. Irradiation hardening of F82H at 573 K and 5 dpa raises the tensile stress to 700-800 MPa, but these specimens only experienced irradiation up to nominally 3.7 dpa.

The laser profilometer measurements were performed in the center 15.24 mm (0.6 inches), with a 7.2° rotation and a 30.48 μ m (0.0012 inch) step between measurements (helical pattern), giving 500 diameter values along the length with 10 total rotations. The value recorded for each point is an average of 2000 measurements with the profilometer at each location/rotation. For the composite imaging 15-20 images were captured along the specimen length and stitched together. Stitching was performed with Adobe Photoshop to retain image resolution.

A jig was developed for cutting the ends off the PCTs that only put gripping force on the ends of the specimens to keep from crimping the thin-walled areas, which would alter the unpressurized dimensional measurements. With this jig the PCT cutting was successfully completed and the internal thermometry was extracted, and dimensional measurements were performed.

The 0 MPa PCT showed 0% creep strain (as expected). The PCT pressurized at 150 MPa showed a creep strain of ~0.18%. The results from the 0 MPa and 150 MPa PCTs are plotted versus other historical data [2] in Figure 3. The creep strain from this work, at the 573 K target temperature, is higher that the creep strain from samples at the same temperature. This possibly due to one (or more) of the following: 1) the irradiation temperature in this work was higher than 573K, 2) the creep mechanism has a dose rate dependence, and/or 3) an affect due to the presence of thermal neutrons. The historical data is from experiments performed at ORNL in the HFIR, but the irradiation was performed in the HFIR reflector where the neutron flux is half of the flux the capsules in this work experienced, and the capsules had a thermal neutron shield, which can alter the rate of helium production and transmutation in the base material.



Irradiation creep data for F82H up to 5dpa

Figure 3. Comparison of creep results from this work with historical measurements presented in [2].

The irradiation capsules contained three temperature monitors, one located inside the PCT and two in the PCT holder. The temperature monitors were removed from the capsule and the PCT during disassembly in the hot cells, and then shipped to the LAMDA for analysis. The determination of the temperature monitor irradiation temperature follows the continuous dilatometer method [3], which determines the irradiation temperature from the annealing behavior of the silicon carbide temperature monitors. The two holder temperature monitors from each irradiation capsule were annealed and analyzed via this method. An example of the dilatometer data and analysis for temperature monitor 16-02 from the FHC01 capsule is given in Figure 4 and for 16-07 from the FHC04 capsule in Figure 5. It is worth mention that all the TMs from FHC01 and FHC02 had similar curve shapes/locations, as did the TMs from FHC03 and FHC04. The results from the TM analysis are listed in Table 1, these results show that the analysis temperature from the TMs within a capsule were identical (accounting for statistical variability) and the TM from FHC01 and FHC02 pair, and FHC04 pair, were within 10-20°C. The last column in Table 1 is the numerical average of the maximum and minimum temperatures, for the TMs from a single capsule.



Figure 4. Continuous dilatometer analysis [3] of TM 16-02 from FHC01 PCT capsule (note behavior is representative for TMs from FHC01 and FHC02).



Figure 5. Continuous dilatometer analysis [3] of TM 16-07 from FHC04 PCT capsule (note behavior is representative for TMs from FHC03 and FHC04).

 Table 1. Summary of continuous dilatometer analysis [3] of PCT TMs. Information on each temperature designation given in [3].

Cansula	Ноор			Maximum	Median	Minimum	Transition	Average
ID	Stress (MPa)	Location	TM ID	Temperature (°C)	Temperature (°C)	Temperature (°C)	Temperature (°C)	Temperature (°C)
	0		16-01	320.8	299.4	266.4	396.1	205 5
FICUI	0	ПКП-Э	16-02	315.1	296.9	279.5	386.7	295.5
ELICON	150		16-03	330	311.3	290	400.6	212.0
FICUZ	150	ПКП-Э	16-09	337.2	320.3	298.4	399.4	313.9
ELICOS	200	ттоц о	16-05	252.9	235.5	210.7	317.5	224.2
FICUS	300	IIKH-3	16-06	254	236.4	219	323.2	234.2
		ттоц 2	16-07	259.3	244	231.1	321.6	228.0
F11C04	300		16-08	251.1	233.4	214.1	319.3	230.9

Once the TM analysis is complete, the results are compared with the TM temperature from the thermal models developed during the design phase. The net difference between the measured/calculated (last column in Table 1) and modeled TM average temperatures is then used to adjust the expected average PCT temperature. For example, if the TM average temperature was 15% higher than designed, then the PCT temperature is also expected to be 15% higher than designed. The summary of the average PCT and SiC TM temperatures from the thermal design, the measured average SiC TM temperatures and the percent difference from the model, and the expected average PCT temperature are listed in Table 2.

Rabbit Number	Average PCT Design Temperature (°C)	Average SiC Design Temperature (°C)	Average SiC TM Measured Average (°C)	% Difference from Average SiC Design Temperature	Expected Average PCT Temperature (°C)
FHC01	300	259	295.5	14.1%	342
FHC02	300	259	313.9	21.2%	364
FHC03	297	259	234.2	-9.6%	269
FHC04	297	259	238.9	-7.8%	274

Table 2. Summary of thermal design results and expected PCT temperature

The two PCTs that retained gas both had temperatures that were ~50°C higher than the target temperature of 300°C, while the two failed specimen temperatures were around 30°C cooler than designed. The cooler temperature of the failed specimens is not surprising because once the specimens failed the pressurization gas will expand to the gas volume in the capsule, including the gas gap incorporated in the design to obtain the target temperature. This will increase the gas pressure within the thermal gas gap, which then allows more heat transfer from the PCT to the holder, housing, and reactor coolant resulting in an overall cooler capsule and PCT. Also, during disassembly, the failed PCTs were stuck inside the holders, suggesting that there was better contact between the PCT the corrugated foil for PCT centering and the holder, which would allow for additional heat transfer away from the PCTs providing further temperature reduction. The capsules all had the same design so it is expected that FHC03 and FHC04 would have experienced the same 350°C irradiation temperature at the start of irradiation as FHC01 and FHC02, if the specimens did not fail prior to the start of irradiation.

The question that remains to be answered is whether the analysis of the SiC TMs can lend insight into whether the failed PCTs lost pressure before irradiation, at low irradiation dose, or near the end of irradiation. The fact that the housings of the failed PCT capsules retained the gas from the failed PCTs suggests that burst of the PCTs occurred after the capsule was assembled and welded shut. This reduces the possibility that the PCTs failed prior to insertion into HFIR. In the temperature range of these capsules, 200-300°C, the radiation swelling in SiC at a given temperature saturates around 1 dpa [4] which is equivalent to half of a HFIR irradiation cycle (12-13 days). Therefore, if the failed PCTs were still retaining pressure after half of the first irradiation cycle, we should expect to observe some annealing of radiation damage at higher temperatures. In other words, the red and purple data sets in the continuous dilatometer analysis should not begin to stabilize until a higher temperature. The two continuous dilatometer sets, shown in Figure 4 and Figure 5, are shown side-by-side in Figure 6 to better highlight the differences in the annealing behavior. The PCTs that retained pressure (FHC01 and FHC02) all had annealing that began to stabilize once the annealing reached 500°C (Figure 6a) whereas the TMs from PCTs that failed (FHC03 and FHC04) all had annealing behavior that stabilized around 400°C (Figure 6b). This behavior suggests that the PCT failures either occurred before irradiation started or early in the first irradiation cycle, otherwise there should have been some SiC annealing occurring at temperatures above 400°C.



Figure 6. Comparison of TM annealing results for (a) FHC01 and (b) FHC04, to enhance comparison of the annealing behavior between a survived and failed PCT.

Future Plans

The future work includes in-depth analysis of dimensional change of all specimens, further investigation as to the temperature results, and authorship of a journal article discussing the results from this work. Microstructural characterization and other property evaluation (like microhardness measurements) may follow if warranted by results of the creep measurements.

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11.2 MINIATURE MECHANICAL TEST DEVELOPMENT FOR TUNGSTEN-BASED MATERIALS—L. M. Garrison, S. K. Wonner, N. C. Reid (Oak Ridge National Laboratory)

OBJECTIVE

The aim of this work is to develop miniature mechanical test methods that can be used to evaluate neutronirradiated tungsten and tungsten composite samples in LAMDA.

SUMMARY

Three-point bending will be used to observe failure modes in tungsten-based samples due to flexural stresses. Because of its simplicity and economy, it is a fitting test to perform on samples of TEM disc geometry that have been irradiated in HFIR. The test is conventionally performed on beams of rectangular cross sections. This test has been adapted to be performed on disc specimens at the plane of symmetry in the center of the sample. The maximum tensile and shear stresses in the sample have been determined for measurements of load and deflection in a universal testing frame.

PROGRESS AND STATUS

Inspiration for the three-point disc bend test is taken from literature and is shown in Figure 1 [1]. In addition, the specimen failure mode can be evaluated via Scanning Electron Microscopy. This technique is useful to measure irradiation induced embrittlement [2]; thus, current work is being done to design a fixture in order to use in these tests. In addition, a mathematical model has been used to relate load-deflection curves from the test frame into stress-strain diagrams in bending.



Figure 1. Three-point bend test schematic, where the distance between the two pins is 2 mm, the total width of the sample is 3 mm, and the radius of curvature for the supports is 0.25 mm [1].

The procedure for use of a three-point bending test in a LAMDA testing frame was adapted from ASTM D790 as well as ASTM E855 [3,4]. A CAD model was developed to be a visual representative of what a test performed in the vacuum furnace testing frame might look like in Figure 2.



Figure 2. CAD model of a three-point bend fixture assembled in the LAMDA C-zone testing frame.

The three-point bend test is used frequently to define material properties, but the real advantage of the test is analyzing the flexural properties in unconventional materials, such as laminates. Thus, fracture mechanisms such as interlaminar failure should be evaluated against the flexural strength and SEM analysis. Flexural strength can be determined by means of calculating the maximum normal stress in tension at the midpoint of the beam.

Due to symmetry, stress is calculated starting from the support at x = 0 to half of the span length, x = L/2:

$$\sigma(x) = \frac{M(x)}{S(x)} = \frac{Px}{2} \cdot \frac{6}{h^2 b(x)} = \frac{3Px}{h^2 b(x)}$$

where $b(x) = \sqrt{4x(L-x) + D^2 - L^2}$,

- *M* is the bending moment in N-mm,
- *S* is the section modulus in mm³, related to the second moment of area,
- *P* is the load in N,
- *x* is the distance from the support in mm,
- *h* is the thickness of the cross-section in mm,
- *b* is the width of the cross-section in mm,
- *L* is the span between both supports in mm,
- *D* is the maximum width of the cross-section, i.e. diameter of the disc, in mm.

For a TEM disc where D = 3, L = 2, h = 0.25:

$$\sigma(x) = \frac{48Px}{\sqrt{(2x+1)(5-2x)}}$$

Figure 3 shows this stress distribution from the support to midpoint of the disc where the load is applied. Specifically, it is the stress at the bottom plane of the sample and is greatest underneath where the load nose is applied at x = 1.0. The stress at this point is equal to 16P and is the flexural strength of the sample where the load is highest. Since disks may be of varying thicknesses, it may be of interest to have dependence of maximum values as a function of thickness, h. In this case, the stress would equal P/h².

The strain at the location of maximum stress is given by the equation:

$$\epsilon = \frac{6\nu_f h}{L^2}$$

Where v_f is the flexural deflection at the midpoint of the disc, as measured by the test frame crosshead displacement or deflectometer. The strength can be reliably determined up to a maximum normal strain of 5%.



Figure 3. $\sigma(x)$ for a varying load. White color refers to a maximum, red a minimum.

Since the span-to-thickness ratio of 8:1 (for h = 0.25) is smaller than the value recommended by ASTM D790 of 16:1, the effects of shear deformation have a stronger effect on the outcome of the test. The shear stresses are highest at the supports and at the neutral axis of the sample due to the non-uniformity of the sample width b, as shown by the equation:

$$\tau(x) = \frac{3P}{4hb(x)}$$

Using the same TEM disc geometry, the shear stress distribution at the neutral axis shown in Figure 4 is:

$$\tau(x) = \frac{3P}{\sqrt{(2x+1)(5-2x)}}$$



Figure 4. $\tau(x)$ for a varying load. White color refers to a maximum, red a minimum.

The shear stress at the neutral axis for different thicknesses is equal to $3P/4h\sqrt{5}$ at the supports and P/4h at the midpoint. For small thicknesses, the stresses in bending are much greater. For the case of a thickness of 0.25 mm, the maximum normal stress is 16 times that of the shear stress at the center of the sample.

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11.3 TENSILE TESTING OF STEEL AND TUNGSTEN FIBERS FOR APPLICATIONS IN COMPOSITE MATERIALS—L. M. Garrison, M. Gussev (Oak Ridge National Laboratory)

OBJECTIVE

The goal of this project is to develop a technique for the tensile testing of steel and tungsten fibers to determine their mechanical properties for use in fiber composites.

SUMMARY

A method for tensile testing steel and tungsten fibers was developed in order to understand the properties of fiber composite materials for use in fusion reactors. A new fixture was designed for tensile testing fibers. To cushion the sample in the fixture, each fiber must be precisely glued into a paper frame. After initial tests, the preparation method was updated. Then, a second round of tests with digital image correlation (DIC) were completed on tungsten and steel fibers.

PROGRESS AND STATUS

Introduction

Tungsten fiber composites are being examined as a possible way to improve the toughness of bulk tungsten materials. The fibers are able to dissipate the crack energy in the composites, could increase their ductility, and lower their effective brittle to ductile transition temperature. In order to understand the mechanical properties of the tungsten fiber composites, it is important to understand the properties of the fibers that will be used to fabricate them. To accomplish this, a method for tensile testing of steel and tungsten fibers is being developed. This involved developing a new fiber testing fixture and procedure. The process of developing a tensile testing technique for the fibers was divided into three tasks. The first was the development of the fiber tensile fixture, second was the development of the fiber sample preparation procedure, and third was the development of the fiber testing procedure. Initial tests were completed on steel and tungsten fibers, as reported in the last semi-annual, and then the procedure was updated and fibers re-tested, as reported here.

Analysis of initial results

It was determined from initial tests that the steel fibers were more prone to pull out during tensile tests than the tungsten fibers. The measured ultimate tensile strength of the steel fibers (~620 MPa) is over four times less than that of the tungsten fibers (~2800 MPa), so if the epoxy is strong enough to hold the tungsten fibers until fracture, it should certainly be strong enough to hold the steel fibers till fracture. It was observed that the steel fibers experience elastic deformation along the entire length of the fiber because there is no thinner gauge section as in more typical tensile tests. In contrast, the tungsten fibers only had deformation in a localized area outside the epoxy. Thus, once the steel fiber began to elastically deform, Poisson's ratio dictated that the fiber dimensions reduce perpendicular to the tensile direction. Once the diameter of the steel fiber was reduced, it would begin to slip out of the epoxy, which did not occur for the tungsten fibers. The sample preparation had to be modified for the steel fibers. Additionally, during preparation of the first set of samples, the epoxy sometimes adhered to the exposed gauge section of the samples, which invalidated the sample, so a new way of applying the epoxy was used.

Modified sample preparation

First the sample testing papers were printed and cut out so that the fiber samples could be sealed between them. Once the paper sample holders had been prepared, the epoxy for attaching the fiber samples to the paper was prepared. The epoxy used was the UHU PLUS Endfest 300. The hardener and binder were first mixed according to manufacturer's directions in a 1:1 ratio by weighing the components on a balance. After the epoxy was thoroughly mixed, it was poured and scraped into a small zip top plastic bag, trying to

minimize the air bubbles in the bag. A small cut was made across one corner of the plastic bag so that the epoxy could be precisely applied on the paper frames, analogous to decorating a cake with a bag of frosting. The use of the small bag of epoxy instead of spreading the epoxy with the plastic tool resulted in much less epoxy on the sides of the samples and outside the paper frames. It also made handling the epoxy much easier and cleaner so there was less transferred to gloves and other tools used in the process. This new method of epoxy application was beneficial for the steel and tungsten fibers.

The other change to the preparation method was specifically for the steel fibers. Using round nose pliers and tweezers, a small bend was introduced in each end of the steel fiber (Figure 1). Care was taken when handling the fibers to only hold near the end and make sure the bend was localized to the end of the fiber. The bends were made in the same plane as much as possible to avoid any torque on the fiber. After the bends were made, the entire straight length of the fiber between the two end bends was measured so that it could be considered the gauge length. When glued into the paper frame, the bend is in the epoxy and helps hold the fiber in place during the tensile test.





Second round of results

Two samples of each of three types of tungsten fibers were tensile tested with DIC. Several of the newly mounted steel fibers were tensile tested without DIC to confirm that the mounting method worked. The special fiber fixture in the tensile frame is shown in Figure 2. The tungsten fiber tests were successful, and the break location was captured in the camera view. That data is being analyzed. The steel fibers with the new mounting method were successful, so two tests of the steel fibers with DIC will also be conducted to conclude this part of the experiment. Next, the fixture and DIC method will be used on the neutron irradiated tungsten fibers.



Figure 2. Steel sample tensile test setup prior to testing before the cutting of the paper support strips.

11.4 HFIR-MFE-RB-19J NEUTRON FLUENCE MEASUREMENTS AND RADIATION DAMAGE CALCULATIONS (REVISED)—L. R. Greenwood, B. D. Pierson, T. Trang-Le (Pacific Northwest National Laboratory)

REVISION AND CORRECTION

This report replaces the previous one published under the same title in Fusion Materials Semiannual Progress Report for the Period Ending December 31, 2018, DOE/ER-0313/65, published in March 2019. The previous version contained incorrect technical information due to communication error.

OBJECTIVE

Measure the neutron fluence and energy spectra during irradiations in HFIR and calculate fundamental radiation damage parameters including dpa and gas production.

SUMMARY

The MFE-19J experiment was irradiated in the removable beryllium (RB) position of HFIR with a 1 mm thick Gd liner [1]. The experimental assembly was irradiated for cycles 466 through 469 starting on June 14, 2016 and ending December 9, 2016 for a total of 8001 MWD at a nominal power of 85 MW. Neutron dosimetry monitors fabricated by PNNL were inserted in the 19J experimental assembly at various elevations. After the irradiation, the neutron fluence monitors were recovered and sent to PNNL for analysis. The activated monitor wires were analyzed to determine activation rates that were used to adjust the neutron fluence spectrum at each irradiation position. The adjusted neutron fluence spectra were then used to calculate radiation damage parameters.

PROGRESS AND STATUS

Neutron dosimetry monitors were fabricated by PNNL for insertion in the 19J experimental assembly. The vanadium monitors measure 0.050" in diameter by 0.340" long. Each monitor has a 2-digit code stamped on the bottom to uniquely identify them. The monitors were filled with small wire segments of Fe, Ti, Nb, and 1% Co-V alloy. Each monitor was electron beam welded to maintain integrity throughout the irradiation. Following irradiation, staff at ORNL removed the monitors from the 19J assembly. The irradiated monitors were received at PNNL for analysis in October 2018. The identities of the monitors, as listed in Table 1, were verified using the 2-digit ID stamp on the bottom. The height above reactor centerline has an uncertainty of \pm 0.7 cm. Monitors 2U and 5H were received and identified for analysis. Unfortunately, the item that was received as monitor 8A was found to not be one of our vanadium monitors and appeared to be a solid metal cylinder, tapered on one end, with a low level of activity compared to the activities in our capsules.

Table 1.	19J	neutron	fluence	monitors
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Monitor	Height, cm
8A	-10.28
2U	2.71
5H	15.21

The monitors were gamma counted as received. The monitors were observed to have some external contamination and they were wiped with wet swabs to reduce the contamination level for further handling. A smear of one of the sample cans was gamma counted and showed activities of ⁶⁰Co, ⁵⁴Mn, ¹³⁴Cs, ¹³⁷Cs, ¹⁵²Eu, ¹⁵⁴Eu, ¹⁵⁵Eu, ⁴⁶Sc, ²⁴¹Am and a few weaker activities. The vanadium monitors were then opened using tubing cutting pliers and the individual wires were retrieved and gamma counted separately in order to look for possible interferences as well as to detect weaker nuclides that could not be detected in the whole monitor counts due to the high level of ⁶⁰Co activity.

For both the 2U and 5H monitors, the ⁶⁰Co activity in the as received monitor was overwhelmingly due to one of the retrieved wire or monitor segments, confirming that the activity was due to the 1% Co-V wire. The ⁶⁰Co activities on other wires or monitor segments were found to be at least a factor of 1000 lower than the activity of the single wires. The niobium wires were retrieved and weighed to confirm the identification prior to processing for x-ray analysis as described below.

The iron wire was retrieved from capsule 5H and the weight was consistent with the pre-irradiation weight such that we could confidently determine the ⁵⁴Mn activity from the iron wire. For monitor 2U, microscopic examination of the retrieved wires showed some evidence of melting as shown in Figure 1. We cannot determine the composition of the melted wire segment 2U but note that the melting points of the materials contained in our monitors range from 1538 C for iron to 3287 C for titanium. The 2U monitor was located between the 800 C and 1100 C regions of the 19J assembly whereas the 5H monitor was between the 1100 C and 550 C regions. The 2U vanadium monitor did not show any signs of melting. We cannot determine if the iron wire melted or perhaps some other wire melted onto the surface of the iron wire since this wire segment also showed lower activities of ⁶⁰Co and ⁴⁶Sc. The ⁶⁰Co activity is produced from the decay of ⁵⁹Fe to ⁵⁹Co and subsequent neutron capture to ⁶⁰Co. However, the ⁴⁶Sc is evidence of some mixing of the iron and titanium wires. The 2U iron wire segment shown in Figure 1 was weighed and the ⁵⁴Mn activity was calculated to be consistent on a per gram basis with the ⁵⁴Mn activity in a larger monitor section thought to contain the entire iron wire. The remaining ⁵⁴Mn activity seen on the as received 2U monitor was due to exterior contamination on the outside of the monitors.



Figure 1. Picture of the iron wires from monitor 5H (left) and 2U (right). Note that the 2U wire segment appears to show evidence of melting.

The titanium wires were not cleanly retrieved. The ⁴⁶Sc activity was found to be present in all the wires or segments removed from the vanadium monitors. For the 5H monitor the sum of the ⁴⁶Sc activities in all the separated wires was used to determine this reaction rate. However, the precision of the detected ⁴⁶Sc in the count of the entire monitor was poor due to the very high ⁶⁰Co activity. For capsule 2U, the ⁴⁶Sc activity could not be cleanly separated from the ⁶⁰Co activity such that this reaction rate could not be accurately determined. This is not unexpected given that the irradiation ended in December 2016 and the ⁴⁶Sc activity has decayed by more than eight half-lives, making it more difficult to detect ⁴⁶Sc in the presence of longer-lived and radiologically intense activities of ⁶⁰Co, ⁵⁴Mn, and ⁹⁴Nb.

The nuclear reactions and activation products that were detected are listed in Table 2. The "90% neutron energy range" represents the energy range of the neutrons that produced 90% of the listed reaction; 5% of the activity was produced by neutrons with energies below the lower energy and 5% above the higher energy. The different activation energy ranges are very useful in defining the neutron spectrum.

Nuclear Reaction	Half-life	90% Neutron Energy Range, MeV
⁵⁴ Fe(n,p)⁵⁴Mn	312 d	2.0 to 8.2
⁴⁶ Ti(n,p) ⁴⁶ Sc	83.8 d	3.7 to 11.0
⁹³ Nb(n,n') ^{93m} Nb	16.1 a	0.66 to 5.0
⁵⁹ Co(n,g) ⁶⁰ Co	5.27 a	5.5E-7 to 1.4E-4
⁹³ Nb(n,g) ⁹⁴ Nb	20,300 a	3.0E-5 to 6.6E-2

 Table 2. Nuclear reactions with energy sensitivity ranges from STAYSL_PNNL calculations

The gamma detectors are calibrated using NIST traceable standards obtained from Eckert and Zeigler. Control counts are performed, every day that a detector is used to check the continuing calibration of the energy, efficiency, and resolution. All nuclear data were adopted from the Nudat 2 database at the National Nuclear Data Center at Brookhaven National Laboratory [2]. The weights of the individual wires were measured during fabrication and all results were reported as activity per gram of the elements with natural abundances using the pre-irradiation masses. The measured activities in Table 3 were corrected back to the end of irradiation time (December 9th, 2016). The activities were corrected for nuclear burnup and gamma absorption to determine reaction rates using the SigPhi calculator from the STAYSL_PNNL software suite [3].

Table 3. Measured activities in Bq/g for the 19J irradiation; uncertainties are ± 2% except for ⁴⁶Ti at ±5%.Results are corrected to the end of irradiation time December 9, 2016. Corrections for decay
during irradiation are included with the activation rates in Table 4.

ID	Height,cm	⁵⁴ Mn	^{93m} Nb	⁹⁴ Nb	⁶⁰ Co	⁴⁶ Sc
		Bq/g	Bq/g	Bq/g	Bq/g	Bq/g
2U	2.71	1.45E+09	2.34E+09	2.59E+07	1.34E+10	*
5H	15.21	9.22E+08	1.69E+09	1.66E+07	6.98E+09	4.88E+08

*Fluence wire not recovered

The ^{93m}Nb activities were determined by x-ray counting of the 16.6 and 18.6 keV x-rays using a low energy photon spectrometer (LEPS) detector. The high purity niobium wires were dissolved in a mixture of nitric and hydrofluoric acid using small Teflon beakers. A small aliquot (about 0.5%) of the solution was accurately weighed and dried on thin filter paper covered by 0.25 mil Mylar for LEPS counting. The very small mass and thin cover nearly eliminated concerns over x-ray absorption, backscatter, and fluorescence as discussed in ASTM practice E1297 [4].

The saturated reaction rates are calculated from the measured activities in Table 3 by applying corrections for decay during irradiation, gamma absorption, and nuclear burnup of the target and product nuclides. These corrections are included with the BCF and SIG-PHI Calculator modules of the STAYSL_PNNL computer suite. (2) The irradiation history was provided by ORNL and is described briefly in the Summary section of this report. The BCF program breaks the irradiation up into periods of nearly constant reactor power and calculates the growth and decay of each activation product. Gamma absorption in the small wire segments is based on the XCOM database (see ref. [5]) as described in section 7.3 of ref [3]. Neutron burnup corrections for thermal neutron reactions use an iterative procedure using a ratio of the burnup reaction rates for the target and product nuclides. The uncorrected reaction rate is used to determine a correction and this procedure is iterated until convergence is achieved. The resulting thermal reactions using known thermal neutron cross sections and resonance integrals. For complete details regarding the iterative neutron burnup correction see figure 28 of ref. [3]. The calculated saturated reaction rates are listed in Table 4.

ID	Height,cm	⁶⁰ Co	⁵⁴ Mn	⁹⁴ Nb	⁴⁶ Sc	^{93m} Nb
		± 2%	± 2%	± 2%	± 10%	± 5%
2U	2.71	4.08E-09	1.35E-11	4.68E-10	*	3.31E-11
5H	15.21	2.11E-09	8.60E-12	2.97E-10	1.20E-12	2.39E-11

Fable 4. Saturated reaction rates in	product atoms/(target atom - second
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*data not usable

The saturated reaction rates are equal to the integral of the neutron activation cross section times the timeaveraged neutron flux over all neutron energies. At each irradiation position we thus have at most 6 integral equations that are solved simultaneously using a generalized least-squares procedure in the STAYSL PNNL computer suite. The neutron activation cross sections were adopted from the IRDFF database compiled by the Nuclear Data Section of the International Atomic Energy Agency [6]. The input neutron spectrum was provided by Charles Daily at ORNL (see Figure 2, blue line) [7]. The input to STAYSL PNNL consisted of the measured saturated reaction rates in Table 4, the input neutron spectrum, the irradiation history, and the IRDFF neutron activation cross sections including all known uncertainties and covariances. Matrix inversion was then used to determine the adjusted neutron flux spectrum and uncertainties including the complete cross-section covariance matrices and a Gaussian formalism-based flux covariance matrix. The results are presented in Table 5 and the neutron spectral adjustments are shown in Figures 2 and 3.

ID	Height	Thermal		Epithermal		> 0.10 MeV		> 1 MeV	
	cm	< 0.5 eV*	±%	0.5 eV to 110 keV	±%		±%		±%
2U	2.71	1.01E+20	28	5.49E+21	6	3.62E+21	6	1.37E+21	5
5H	15.21	5.83E+19	28	3.35E+21	6	2.44E+21	6	9.38E+20	5

 Table 5. Neutron fluence values (n/cm²) from the STAYSL_PNNL spectral adjustment

*Due to the Gd liner, the thermal fluence is very sensitive to the energy threshold. A higher threshold of 2.1 keV is shown later in Table 7 and Figure 4.



Figure 2. Neutron spectral adjustment with STAYSL PNNL for the 2U position at 2.71 cm. Flux per lethargy is shown both before and after adjustment. The dashed curve shows the size of the adjustment using the percent difference scale on the right side.



Figure 3. Neutron spectral adjustment with STAYSL PNNL for the 5H position at 15.21 cm. Flux per lethargy is shown both before and after adjustment. The dashed curve shows the size of the adjustment using the percent difference scale on the right side.

Radiation Damage Calculations

The adjusted neutron spectrum determined at each positon in the irradiation assemblies was used to calculate the dpa and helium production in various elements and alloys using the SPECTER computer code [8] and the damage parameters are listed in Table 6. The radiation damage calculations do not take into account nuclear transmutation that occurs during irradiation. In most cases the transmutation does not significantly change the dpa or helium production but may lead to changes in the alloy composition. Due to the Gd thermal filter in this experiment, transmutation effects are minimal. The dpa and helium values for nickel also include the extra contribution from ⁵⁹Ni. The materials irradiated in the 19J experiment are provided in reference 8. Based on this list, radiation damage calculations were performed for Fe, Ni, W, F82H, and SiC.

Material	2U 2.	71 cm	5H 15.21 cm		
	dpa	He,appm	Dpa	He,appm	
Fe	2.12	0.68	1.43	0.37	
Ni*	2.35	9.62	1.58	5.15	
W	0.58	0.0083	0.39	0.0050	
F82H+	2.11	0.65	1.42	0.38	
SiC	3.81	11.26	2.57	6.64	

Table 6. Radiation damage parameters for the 19J experiment

*Ni dpa and helium values include contributions from ⁵⁹Ni

+F82H composition 8Cr-2W-0.04Ta (balance Fe)

Fluence and Radiation Damage Polynomial Fits and Comparison to ORNL Calculations

The neutron fluence and radiation damage parameters can be well described by a polynomial function. The form of the polynomial is given as equation (1) below where a is the maximum value, and b is the quadratic parameter. The flux, dpa, and helium (appm) values at any location in the assembly can then be determined using the height, z in cm, and the coefficients listed in Table 7. The functions are symmetric about core midplane. The accuracy is limited since we only have two data points for the determination of the parameters. The heights of the monitors above reactor centerline have an uncertainty of \pm 0.7 cm.

$$F = a(1 + bz^2)$$
 (1)

Neutron Fluence	a, n/cm²	b, cm ⁻²		
Thermal < 0.5 eV	1.02E+20	-1.86E-03		
Thermal < 2.1 eV*	4.82E+20	-1.94E-03		
Epithermal	5.56E+21	-1.72E-03		
>0.1 MeV	3.59E+21	-1.44E-03		
> 1 MeV	1.38E+21	-1.39E-03		
Damage parameter	a, dpa	b, cm ⁻²	a, He, appm	b, cm ⁻²
Fe	2.14E+00	-1.44E-03	6.89E-01	-1.81E-03
W	5.86E-01	-1.45E-03	8.41E-03	-1.75E-03
F82H	2.13E+00	-1.44E-03	6.59E-01	-1.83E-03
SiC	3.85E+00	-1.44E-03	1.14E+01	-1.81E-03

Table 7. Polynomial coefficients for Equation 1 to calculate neutron fluence and damage parameters

*Due to the Gd liner, the thermal fluence is very sensitive to the cutoff energy. A higher energy threshold was used for comparison with the MCNP calculations shown later in Figure 4.

Figure 4 compares our fluence measurements with calculations by C. Daily at ORNL [7]. Specimens in the RB-19J assembly were placed in vertical holes as documented in reference 1. Fast flux calculations were provided by C. Daily for 7 holes around the outside of the assembly. The fast fluence decreases with radial distance from core center as shown in the figure. To reduce this effect, the assembly was rotated between each of the four reactor cycles. These calculations were thus averaged to determine the fluence values shown in Figure 4. The PNNL measurements and uncertainties are shown by the larger red dots. The height of the fluence monitors is known to within 0.7 cm. The PNNL epithermal and fast neutron fluence measurements at 15.2 cm agrees well with the calculation and the measurement at 2.7 cm is within 2-sigma uncertainties. The PNNL thermal fluence measurements have high uncertainties due to the Gd filter.



Figure 4. Comparison of measured (PNNL, red) and calculated (ORNL, blue) neutron fluence values for the epithermal fluence between 0.5 eV and 76 keV, fast fluence > 0.1 MeV and the thermal fluence < 2.1 eV. The dotted red lines are polynomial fits to the PNNL fluence measurements and ORNL calculations.

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