FEB 1 2 1991

PNL-SA--19062

DE91 007200

RADIATION-INDUCED CHANGES IN ELECTRICAL CONDUCTIVITY OF A WIDE RANGE OF COPPER ALLOYS

FA Garner KR Anderson(a) T. Shikama^(b)

January 1991

To be included in the Fusion Reactor Materials Semiannual Progress Report DOE/ER-0313/9

Work supported by the U.S. Department of Energy under Contract DE-AC06-76RL0 1830

Pacific Northwest Laboratory Richland, Washington 99352

- University of Illinois (a)
- Urbana, Illinois Tohoku University (b) Japan

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States bility for the accuracy. completeness, or usefulness of any information, apparatus, product, or Government. Neither the United States Government nor any agency thereof, nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsiprocess disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, mendation, or favoring by the United States Government or any agency thereof. The views manufacturer, or otherwise does not necessarily constitute or imply its endorsement, recomand opinions of authors expressed herein do not necessarily state or reflect those of the Jnited States Government or any agency thereof.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

MASTER

RADIATION-INDUCED CHANGES IN ELECTRICAL CONDUCTIVITY OF A WIDE RANGE OF COPPER ALLOYS - F. A. Garner, Pacific Northwest Laboratory, (*) K. R. Anderson, University of Illinois, and T. Shikama, Tohoku University

OBJECTIVE

The objective of this effort is to identify those copper alloys that offer promise as high heat flux materials for fusion application.

SUMMARY

A wide variety of radiation-induced changes in electrical conductivity was observed in a series of irradiation experiments conducted on copper alloys in FFTF/MOTA. The behavior of each alloy was found to depend on the alloy composition, starting state, irradiation temperature, and the sometimes complex interaction of three radiation-driven processes. These processes are transmutation, void swelling, and solute redistribution.

PROGRESS AND STATUS

Introduction

A variety of copper alloys have been proposed for high heat flux applications for both near-term and longterm fusion goals. In response to these proposals, a series of exploratory irradiation experiments are being conducted in FFTF/MOTA.⁽¹⁾ One property being measured in this experimental series is the electrical conductivity, a property that often changes substantially in response to the combined influence of void swelling, transmutation, and solute redistribution.

Several generations of FFTF copper irradiation experiments have been conducted, and the latest of these, designated Generation 2.0, continues to accumulate exposure in FFTF. Changes in conductivity of some but not all alloys measured in Generations 1.0, 1.5, and 2.0, have been reported previously, $^{(2,3)}$ and conductivity measurements continue to be performed on alloys that were bypassed in the first series of measurements. Measurements of conductivity for all alloys irradiated in the 430°C Generation 1.0 series at 16, 47, 63, and 98 dpa, the Generation 1.5 experiment at 414°C, 34 dpa and 529°C, 32 dpa, and the 411°C Generation 2.0 experiment at 50 dpa have now been completed and are reported here.

Measurements of the 411°C Generation 2.0 alloys at 100 and 150 dpa will be performed during 1991. Anderson et al.⁽²⁾ present the details of the experimental technique and Garner et al.⁽¹⁾ provide a detailed description of the materials studied.

Some data presented in this report are reevaluations of previously reported data, based on refinements in the measurement technique and involving changes usually on the order of 10% or less. At the higher fluence levels of the Generation 1.0 experiment it was also found to be necessary to remeasure some previously reported specimens after mechanically removing a surface layer of aluminum. This layer had formed on closely packed specimens by diffusion bonding with aluminum spacers inserted between the specimens. ⁽¹⁾ This problem became more pronounced as void swelling caused an increasingly tighter packing of the specimens. After removing the aluminum, a significant improvement was observed in the reproducibility of the measurements and in their consistency with data taken at lower fluence levels.

Results and Discussion

A compilation of the new and revised conductivity data measured in the various generations of the FFTF/MOTA copper irradiation experiments is presented in Tables 1 and 2.

Pure copper has been used as a standard reference material in the various generations of the FFTF copper irradiation program as well as in programs conducted in other reactors. For this purpose, we have chosen to examine the reproducibility of its electrical conductivity in these FFTF studies and in another fast reactor irradiation experiment conducted in EBR-II.⁽⁴⁾ The major transmutation product in copper is nickel and its production rate is very dependent on neutron spectra. Nickel additions to copper have a substantial impact on the electrical and thermal conductivities. Both nickel and the second major transmutant, zinc, strongly depress the conductivities of copper.^(4,5) Void swelling also decreases both types of conductivity.

Figure la shows that fast reactor irradiations of pure copper at 385-430°C yield electrical conductivity changes that are very consistent. Since the neutron spectra of EBR-II and FFTF are very similar, the consistency is not unexpected. Also shown in Figure la is electrical conductivity data for pure copper

⁽a) Operated for the U.S. Department of Energy by Battelle Memorial Institute under Contract DE-ACO6-76RLO 1830.

Table 1.

Electrical Conductivities (%IACS) of Generation 1.0 Copper Alloys Irradiated as TEM Disks in FFTF/MOTA at -430°C

Material Alloy	Code	0 dpa	16 dpa	47 dpa	63 dpa	98 dpa
MARZ Cu (SA)	RO	100.9	88.0	61.1	55.0 ^(b)	32.7 ^(b)
Cu-5Ni (SA)	R7	29.2	23.0 ^(a) 25.0 ^(a)	-	19.8	13.9 ^(b)
CuBe (HTB)	R3	20.2 ^(a)	32.0(*)	32.6 ^(b)	$29.1^{(a)}$	32.8 ^(b)
CuBe (IITA)	R1	19.3 ^(a)	31.1 ^(a)	29.2 ^(a)	-	27.3 ^(b)
CuNiBe (HTB)	RT	54.5 ^(a)	64.0 ^(a)	54.2	45.7 ^(b)	-
CuNiBe (HTA)	RR	66.8 ^(a)	75.9 ^(a)	53.0	47.3 ^(b)	24.1 ^(b)
CuAg (SA)	RX	94.1 ^(a)	74.5 ^(b)	46.6	45.2 ^(b)	29.0 ^(b)
Cu∧gP (SA)	RV	86.4 ^(a)	71.9 ^(a)	42.8	-	27.5 ^(b)
MZC (CWA)	RU	83.3 ^(a)	77.1	59.6	-	-
Cu-5Al (SA)	R9	(c)	27.7	-	-	12.6 ^(b)
CuAl25 (CW)	R4	85.5 ^(a)	-74	-50	~61 ^(b)	36.4 ^(b)
CuA125 (CWA)	R5	79	-	-	~59 ^(b)	58.2 ^(b)

(a) Reevaluation of previously published data.

(b) Value measured after removing aluminum-contaminated surface.

(c) Not yet measured.

derived from FFTF irradiation at 529°C. Since the swelling at this dose and temperature is only 1.8% (a factor of ten lower than at -400°C) most of the observed conductivity loss arises from transmutation to nickel and zinc, a process that is independent of temperature.

In pure copper it was found that the effects of transmutation and voidage on electrical resistivity were directly additive in accordance with Matthiesson's rule, indicating that no significant interaction between the two processes has occurred.^(4,5) Two other alloys in the Generation 1.0 study involved relatively low levels of solute and one might expect a similar lack of interaction. These alloys are CuAg and CuAgP. Note in Figure 1b that the addition of solutes in CuAg and CuAgP lowered the preirradiation conductivity from the 101% IACS of pure copper to 94 and 86% IACS, respectively. This difference in conductivity was maintained relative to that of pure copper with little change throughout the first generation experiment, indicating that these solutes probably stayed in solution. The initially low conductivity of Cu-5Ni reflects the influence of nickel and decreases further with irradiation, as is also shown in Figure 1b. Much of the radiation-induced decrease in Cu-5Ni is due to void swelling, the development of which appears to be unaffected by the presence of the nickel.⁽⁶⁾

There are some kinds of nonadditive behavior that can occur, however. In most solute-modified alloys, radiation-induced redistribution of initial solutes also leads to modifications in conductivity, particularly if the solute levels are relatively large. A previous paper⁽⁵⁾ showed that the electrical conductivity of Cu-2.0Be initially increased slightly with irradiation, but remained constant thereafter (see Figure 2). The initial increase might be attributed to radiation-induced acceleration of precipitation and aging, but the constant conductivity thereafter was thought to be inconsistent with the continued accumulation of nickel and zinc via transmutation. It now appears that the transmutant nickel drives some of the beryllium out of solution as it forms, and the two effects compensate. The strongly reduced solubility of beryllium in copper-containing nickel is the principle on which the higher conducting CuBeNi (Cu-0.3Be-1.8Ni) alloy was developed to replace Cu-2.0Be. In CuBeNi, the lower level of beryllium compared to that of Cu-2.0Be leads to a much higher level of initial conductivity. The conductivity is increased slightly by solute redistribution in the early stages of irradiation, but this trend is reversed at a relatively low exposure by accumulating levels of transmutants and swelling. Microstructural observations show that CuBeNi swells relatively easily and that CuBe does not swell; ^(7,8)

Allov -	Mat Code	terial O dpa	34 dpa	414°C 50 dpa	411°C 529°C 32 dpa
MAD7 Cu		100.0	70 4	59.1	02 7(b)
MARZ CU	ĸu	100.9	70.4	58.1	90.8 ^(b)
CuA125	R4	85.7		83.2	
CuA125 (welded)	3N 3N	77.7	81.0	59.6
		73.9		57.7	
CuA120	UX	88.5	81.6	72.7 ^(D)	84.0
CuA120	U7	84.9	82.0	10.3.	00.4
CuA115B	võ	91.6	80.0	74.7(b)	85.8 ^(b)
AD.C. 1			83.6	73.8 ^(b)	88.6(6)
005-1	31	80.6		63./ 59.3	
ODS-1 (w	elded)	3H	88.4	59.5	67.9
(,	88.5		64.9	
0DS - 2	3K	85.7(5)		67.2 ^(b)	1
		88.7 ^(b)		73.6 ^(b)	
005-3	31	86.0		50 (b)	
00.0	52	89.0		57.0 ^(b)	
0DS - 4	3M	79.8		67.4	
~ ~		78.5		72.0	
Lutr	34	89.4		77.9	
Cullf	38	88.8		77.6	
CuNiTi	VK	40.4 ^(b)	52.3	48.7 ^(h)	68.7 ^(b)
		40.9 ^(b)	52.9	47.3 ^(b)	65.0(6)
		37.50		40.2007	
	40.5				
		40.8			
CuNiTi	VL	31.4	56	48.6 ^(b)	$67.2^{(b)}$
		32.9	59	49.80	68,9(0)
CuNiSn	VN	15.8	40.8		
		16.5			
		16.5 (b)			an (h)
		$18.0^{(b)}$			27.9(b)
		18.6 ^(h)			27.7 ^(b)
CuBe (IIT	B)	R3	20.7		31.8
				30.8	
Cu-5N1	٧P	29.2	18.4		
CuNiRe (AT)	U1	58.2	67.9	
currec (,	.	66.3	0,15	
CuNiBe (1/11)	U3	71.0	78.8	66.5
CUNEDA	11.4	73 C	74.8		
CUNTRE	04	13.0	82.5		
MZC	U6	88.2	88.0		90.5
			77.9		91.1
MZC	U7	86.7	88.0		
MZC (MIT	#2)	119	84.3	85.7	81,083,6
			88.6	77.9	86.2
MZC (MIT	#3)	VB	87.3	85.2	80.391.8
			86.1	78.7	90.5

Electrical Conductivities (% IACS)^(a) of Generation 1.5 and 2.0 Copper Alloys Irradiated in FFTF/MOTA

_

(a) Multiple values represent measurements from separate specimens.(b) Indicates TEM disks; no notation denotes measurements made on gage section of miniature tensile specimen.

-1



Figure 1. a) Neutron-Induced Changes in Electrical Conductivity of Pure Copper in Various Fast Reactor Irradiation Experiments in FFTF and EBR-II, b) Changes Observed in Cu-5Ni, CuAg, and CuAgP Irradiated in FFTF.



Figure 2. Wide Variety of Electrical Conductivity Behavior Observed in Five Alloys Irradiated in FFTF, Illustrating the Range of Interactions Between Precipitation , Solute Redistribution, Transmutation, and Void Swelling. The designations HTA, HTB, SAA, CWA denote Cold-Worked, Annealed, Annealed and Aged, and Cold Worked and Aged, respectively.

٦

The complexity and variety of solute-related changes in conductivity for various copper alloys is also illustrated in Figure 2. The large increases in conductivity observed in the CuNiTi and CuNiSn spinodally strengthened alloys are due to the dominant effect of radiation-induced and temperature dependent overaging of precipitates formed by spinodal decomposition, but at higher fluence the influence of void swelling and precipitation eventually begins to overwhelm the influence of solute redistribution and the conductivity begins to decline.⁽⁹⁾

Internally oxidized alumina alloys exhibited the best overall performance in the Generation 1.5 and 2.0 experiments, retaining the largest fractions of their electrical conductivity, $^{(2)}$ strength $^{(10,11)}$ and swelling resistance. $^{(6,10,11)}$ Swelling in this class of alloys was found to be very low, dependent on oxide content, and possibly dependent on cold work level. There is essentially no dependence of the electrical conductivity of these alloys on irradiation temperature since, in the absence of substantial void swelling, transmutation accounts for the majority of the decrease in conductivity. $^{(2,9)}$ The conductivity behavior of this class of alloys was covered in more detail elsewhere. $^{(2)}$ Some of these data are reproduced in Figures 3 and 4 for comparison with the behavior of the solute-strengthened alloys.

Conclusions drawn in this report concerning the origin of the conductivity change of a given alloy are derived not only from microstructural observations but also from changes in mechanical properties and density. (7-9,11-13) At higher exposure levels in alloys that do not resist swelling, microstructural examination is not always feasible, and the behavior must be inferred from changes in the macroscopic properties.









Figure 4. Measurements of Electrical Conductivity of Both Unirradiated and Irradiated (50 dpa at 411°C) Dispersion-Strengthened and Spinodally-Strengthened Alloys from Generation 2.0.

CONCLUSIONS

A wide variety of radiation-induced behaviors was observed in the electrical conductivity of various copper alloys irradiated in FFTF/MOTA. This complexity arises from the sometimes interactive effects of composition, starting state and temperature on three competing processes: transmutation, void swelling, and solute redistribution. The latter process can either increase or decrease the conductivity, while swelling and transmutation in general decrease the conductivity. In some cases these various contributions to conductivity can interact strongly to yield unanticipated results.

FUTURE WORK

Conductivity measurements will proceed on Generation 2.0 alloys irradiated at 411°C to 100 and 150 dpa. Specimens for irradiation in Generation 3.0 are now being prepared.

REFERENCES

1. F. A. Garner, M. L. Hamilton, K. R. Anderson, J. F. Stubbins, B. N. Singh, A. Horsewell and W. F. Sommer, "Overview of Copper Irradiation Programs," <u>Fusion Reactor Materials Semiannual Progress</u> <u>Report</u>, DOE/ER-0313/6 (1989), pp. 351-356.

2. K. R. Anderson, F. A. Garner, M. L. Hamilton and J. F. Stubbins, "Electrical Resistivity Changes Induced in Copper Alloys by Fast Neutron Irradiation," <u>Fusion Reactor Materials Semiannual Progress Report</u>, DOE/ER-0313/9, pp. 357-369.

3. F. A. Garner and K. R. Anderson, "Conductivity Changes Observed in Generation 1.0 Copper Alloys Irradiated at 430°C to 16-98 dpa," <u>Fusion Reactor Materials Semiannual Report</u>, DOE/ER-0313/7 (1989), pp. 220-222.

4. H. M. Frost and J. C. Kennedy, <u>J. Nucl. Mater.</u> 141-143 (1986) 169-173.

5. F. A. Garner, H. L. Heinisch, R. L. Simons, and F. M. Mann, Radiation Effects, 113 (1990) 229-255.

6. F. A. Garner and H. R. Brager, "Neutron-Induced Changes in Density of Copper Alloys," in this semiannual report.

7. II. R. Brager, <u>J. Nucl. Mater.</u> 141-143 (1986) 79-86.

8. H. R. Brager, J. Nucl. Mater. 141-143 (1986) 163-168.

9. K. R. Anderson, "The Properties of Oxide Dispersed and Spinodally Strengthened Copper Alloys Following High Temperature Neutron Environment Exposure", Doctoral Thesis, University of Illinois at Urbana-Champaign, 1970.

10. II. R. Brager, II. L. Heinisch and F. A. Garner, <u>J. Nucl. Mater.</u> 133 & 134 (1985) 676-679.

11. K. R. Anderson, F. A. Garner, M. L. Hamilton and J. F. Stubbins, in this semiannual report.

12. T. Shikama, F. A. Garner, M. L. Hamilton, and K. R. Anderson, "Tensilg and Fracture Behavior of Cu-5Ni and Various Precipitation-Strengthened Copper Alloys After High Fluence Irradiation", PNL-SA-17549, to be included in the Proceedings of the 15th ASTM International Symposium on Effects of Radiation on Materials, Nashville, TN, June 17-21, 1990.

13. H. R. Brager and F. A. Garner, "Effects of Neutron Irradiation to 98 dpa on the Swelling of Various Copper Alloys," ASTM STP 1046 (vol. II), N. H. Packan, R. E. Stoller, and A. S. Kumar, eds. 1990, pp. 599-604.



DATE FILMED

