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Atom Probe Tomography Characterization of the Solute Distributions in a Neutron-Irradiated and Annealed Pressure Vessel Steel Weld

Oak Ridge National Laboratory

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ABSTRACT

A combined atom probe tomography and atom probe field ion microscopy study has been performed on a submerged arc weld irradiated to high fluence in the Heavy-Section Steel Irradiation (HSSI) fifth irradiation series (Weld 73W). The composition of this weld is Fe – 0.27 at. % Cu, 1.58% Mn, 0.57% Ni, 0.34% Mo, 0.27% Cr, 0.58% Si, 0.003% V, 0.45% C, 0.009% P, and 0.009% S. The material was examined after five conditions: after a typical stress relief treatment of 40 h at 607°C, after neutron irradiation to a fluence of 2×10^{23} n m^{-2} (E > 1 MeV), and after irradiation and isothermal anneals of 0.5, 1, and 168 h at 454°C. This report describes the matrix composition and the size, composition, and number density of the ultrafine copper-enriched precipitates that formed under neutron irradiation and the change in these parameters with post-irradiation annealing treatments.

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FOREWORD

The work reported here was performed at the Oak Ridge National Laboratory (ORNL) under the Heavy-Section Steel Irradiation (HSSI) Program, T. M. Rosseel, Program Manager. The program is sponsored by the Office of Nuclear Regulatory Research of the U.S. Nuclear Regulatory Commission (NRC). The technical monitor for the NRC is C. J. Fairbanks.

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INTRODUCTION

The embrittlement of pressure vessel steels during service in a nuclear reactor is clearly evident from the change in mechanical properties. These changes are manifested in an upward shift in the ductile-to-brittle transition temperature and a loss in fracture toughness. These changes have been correlated with the composition of the pressure vessel steel, in particular with the levels of copper, nickel, manganese, and phosphorus, and also with the magnitude of the fluence of the irradiation.

Previous atom probe field ion microscopy investigations of neutron-irradiated pressure vessel steels have clearly established that there are several different types of microstructural features present in these materials. The atom probe field ion microscope has indicated that the most prevalent change in the microstructure during neutron irradiation is the evolution of ultrafine (~2 nm) copper-enriched regions and phosphorus-enriched regions. Other microstructural changes that have been characterized include solute segregation to and precipitation at dislocations and grain and lath boundaries. The contributions of the techniques of atom probe field ion microscopy and atom probe tomography to the understanding of the microstructure of neutron-irradiated pressure vessel steels and related alloys have been reviewed recently [1].

This report presents the results of an atom probe tomography study of the effects of high-fluence neutron irradiation and subsequent short- and long-term post-irradiation annealing treatments of a high-copper submerged arc weld. The technique of atom probe tomography is an extension to atom probe field ion microscopy that enables the x, y, and z coordinates and the elemental identities of the atoms within a small volume to be determined with atomic resolution [2–4]. The analysis volume typically contains between 500,000 and 1,000,000 atoms and originates from a volume in the specimen that is ~10 to ~15 nm square by ~100 to ~250 nm long. These data may then be reconstructed in a computer so that the distribution of all the elements present in the material may be visualized. In addition, material and microstructural parameters such as the size, shape, and number density of ultrafine precipitates and the compositions of precipitates and the surrounding matrix may be estimated.

EXPERIMENT

This atom probe tomography and atom probe field ion microscopy study was performed on a submerged arc weld from the HSSI fifth irradiation series (Weld 73W). The chemical composition of this high copper (0.27 at. % Cu) weld is given in Table 1 [5]. The material was examined after five conditions: after a typical stress relief treatment of 40 h at 607°C, after neutron irradiation to a high fluence of 2×10^{23} n m^{-2} (E > 1 MeV), and after irradiation

Table 1. Bulk chemical composition of the submerged arc weld (weld 73W)

Element	Atomic percent	Weight percent
Cu	0.27	0.25
Mn	1.58	1.56
Ni	0.57	0.60
Mo	0.34	0.58
Si	0.89	0.45
Cr	0.27	0.25
C	0.45	0.098
P	0.009	0.005
S	0.009	0.005
Fe	Balance	Balance

and isothermal anneals of 0.5, 1, and 168 h at 454°C. Blanks (0.25 × 0.25 × 10 mm) for atom probe specimens were cut from Charpy bar specimens with the use of a slow-speed diamond saw. The electropolishing methods used to prepare needle-shaped field ion specimens from these bulk samples have been described previously [6].

The material was characterized in both energy-compensated atom probe field ion microscopes (ECAP) [7,8] and an energy-compensated optical position-sensitive atom probe (ECOPoSAP) [4]. The former type of instrument was used primarily for characterization of the composition of the matrix and for general field ion microscopy of the microstructure. This instrument features high mass resolution. The latter three-dimensional ECOPoSAP instrument was used primarily to provide information on the size, morphology, number density, and composition of the copper-enriched regions formed during irradiation, since the number of atoms collected from each specimen was significantly higher. In both instruments, the conditions used for composition determinations were a specimen temperature of 50K, a pulse fraction of 20% of the standing voltage, and a pulse repetition rate of 1500 Hz. Field ion images were recorded with a specimen temperature of 50K and with the use of neon as the image gas.

RESULTS AND DISCUSSION

The results of the matrix composition determinations for all the materials are summarized in Table 2. It should be noted that these determinations do not include any contributions from the copper-enriched regions and are strictly the copper levels in solid solution in the matrix. As in previous investigations of pressure vessel steels [1], the copper level in the matrix was found to decrease during neutron irradiation, and a small additional decrease in the copper level was observed after post-irradiation annealing. These results are in agreement with previous studies of a surveillance weld and a weld from the Midland reactor [9,10].

Table 2. Matrix compositions of the submerged arc weld (weld 73W) for the five different heat treatments in atomic percent

Element	Stress relieved	Irradiated 2×10^{23} n m^{-2} (E > 1 MeV)	Irradiated + annealed for 0.5 h at 454°C	Irradiated + annealed for 1 h at 454°C	Irradiated + annealed for 168 h at 454°C
Cu	0.12	0.055	0.052	0.049	0.039
Mn	0.94	0.78	1.00	0.82	0.89
Ni	0.51	0.53	0.70	0.57	0.89
Mo	0.15	0.30	0.17	0.19	0.12
Si	0.75	0.62	0.61	0.51	0.66
Cr	0.14	0.20	0.16	0.18	0.15
C	0.01	0.027	0.023	0.01	0.01
P	0.011	0.024	0.003	—	—
Fe	Balance	Balance	Balance	Balance	Balance

— = not detected.

A series of three-dimensional atom maps of the copper, nickel, and manganese atom distributions are shown in Figs. 1–4 for the different materials. In this type of representation, each sphere is the location of an individual atom in the volume [4]. It is evident from the local increase in the density of copper atoms that ultrafine copper-enriched regions are present in all neutron-irradiation and post-irradiation annealed materials. It is also evident from the comparison between the copper, nickel, manganese, and silicon atom maps that the copper-enriched regions are also enriched in nickel, manganese, and silicon. The size and shape of each copper-enriched region may be obtained from these atom maps. The copper-enriched regions were found to be roughly spherical, but the surface of the enriched region was extremely irregular on an atomic scale. The irregular surface has previously been described as a ramified structure [6]. These observations are in agreement with previous atom probe characterizations of neutron-irradiated pressure vessel steels [1].

In addition to the copper-enriched precipitates, phosphorus-enriched regions were also observed. An example of a phosphorus-enriched region in the material that was neutron-irradiated and annealed for 0.5 h at 454°C is shown in Fig. 5. No copper was associated with this phosphorus-enriched region. The number density of these features was at least an order of magnitude lower than that of the copper-enriched regions.

This type of high spatial resolution analysis also enables estimation of the number of atoms associated with each copper-enriched region. Since the spatial coordinates of all the copper atoms are known, the copper atoms that are associated with each copper-enriched precipitate can be determined by locating the copper atoms that were within 0.7 nm of other copper atoms. The value of 0.7 nm was selected from the visual examination of extent of the copper-enriched regions and the absence of the copper concentration in the atom maps. The results for the different materials are summarized in Table 3. It is evident that there is a large variation in the number of copper atoms associated with each precipitate. The minimum size of the precipitate may be estimated from the number of copper atoms by assuming that the copper atoms are arranged within a spherical envelope on a body-centered cubic lattice with the same lattice parameter as iron (or any lattice with the equivalent atomic volume). These results are included in Table 3. It should be noted that these results slightly underestimate the true minimum size, as they are not corrected for the detection efficiency of the mass spectrometer. This correction increases the minimum radius of the precipitate by approximately 12%.

In addition, the center and the radius of gyration of each copper-enriched region can be calculated from the positions of the copper atoms. The center of mass ($\bar{x}$, $\bar{y}$, $\bar{z}$) of a feature, such as a precipitate, is given by

$$\bar{x} = \frac{\sum_{i=1}^{n} x_i m_i}{\sum_{i=1}^{n} m_i}, \quad \bar{y} = \frac{\sum_{i=1}^{n} y_i m_i}{\sum_{i=1}^{n} m_i}, \quad \text{and} \quad \bar{z} = \frac{\sum_{i=1}^{n} z_i m_i}{\sum_{i=1}^{n} m_i} \tag{1}$$

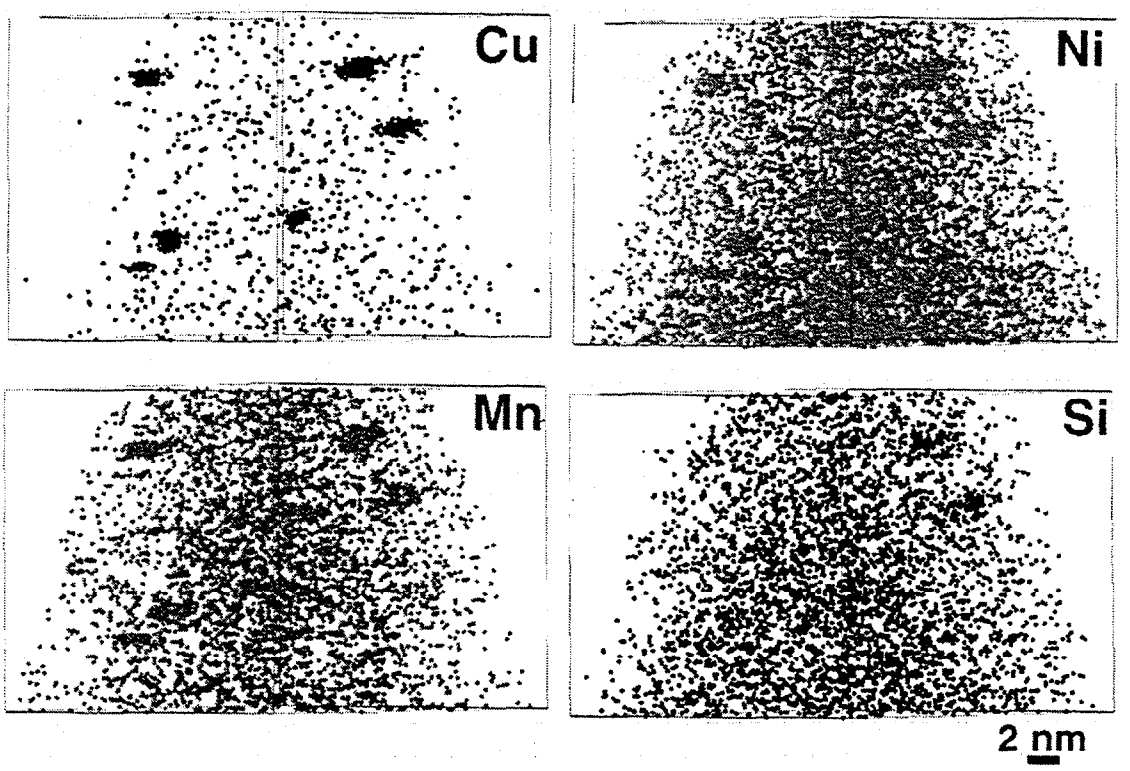

Figure 1. Atom maps of the solute distribution in weld 73 after neutron irradiation to a fluence of 2×10^{23} n m^{-2}.

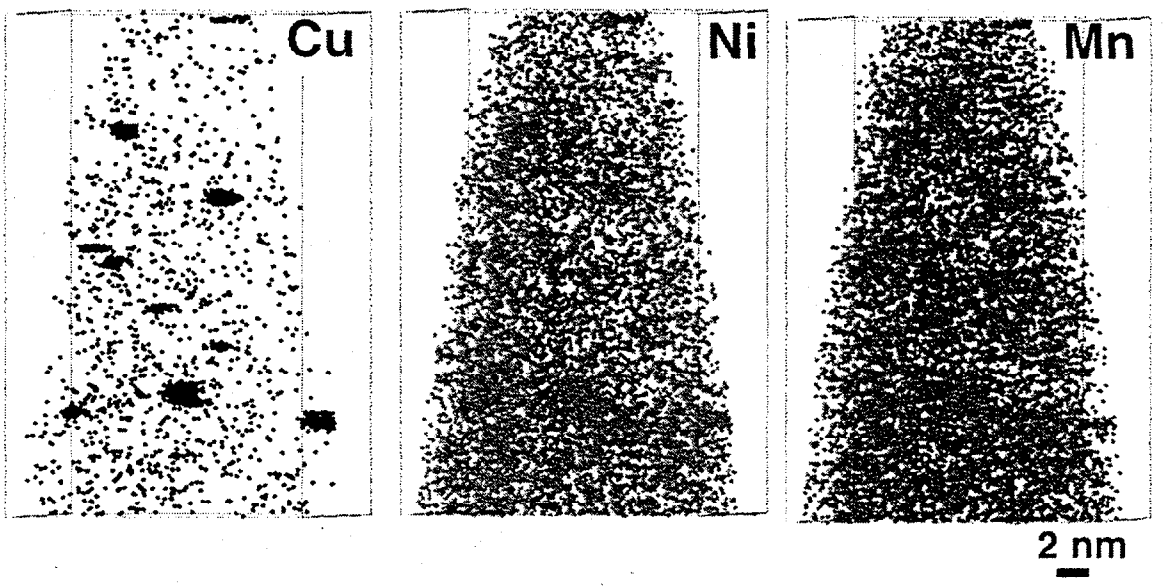

Figure 2. Atom maps of the solute distribution in weld 73 after neutron irradiation to a fluence of 2×10^{23} n m^{-2} and anneal for 0.5 h at 454°C.

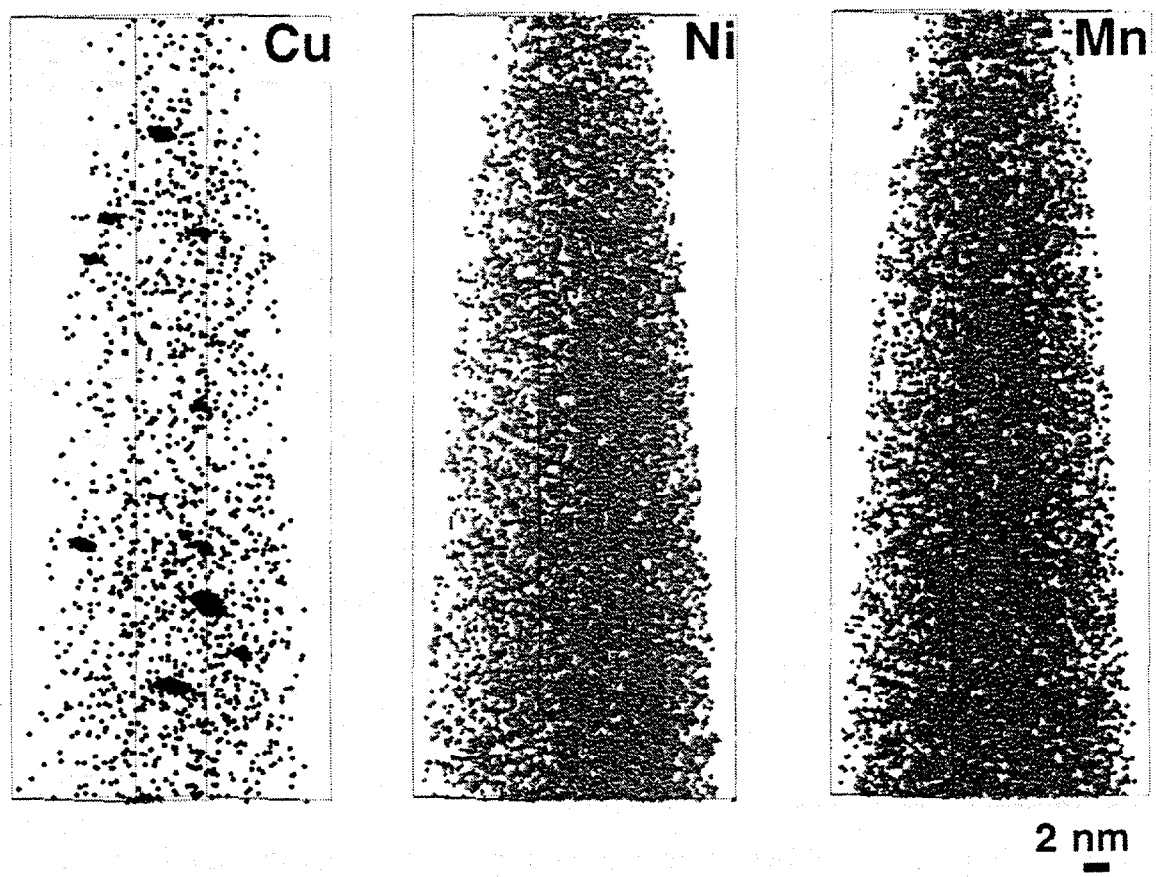

Figure 3. Atom maps of the solute distribution in weld 73 after neutron irradiation to a fluence of 2×10^{23} n m^{-2} and anneal for 1 h at 454°C.

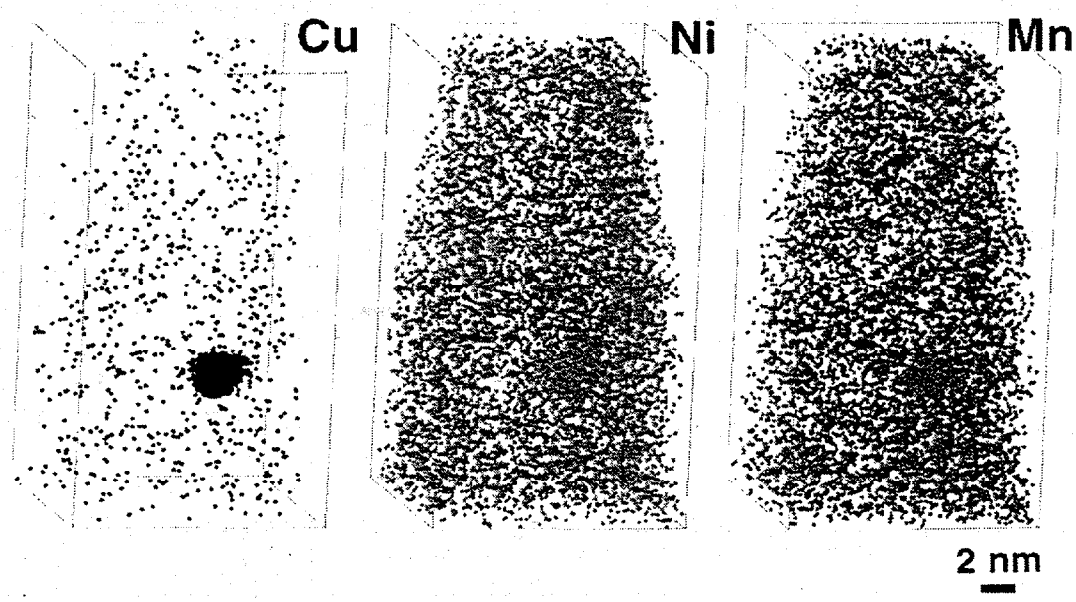

Figure 4. Atom maps of the solute distribution in weld 73 after neutron irradiation to a fluence of 2×10^{23} n m^{-2} and anneal for 168 h at 454°C.

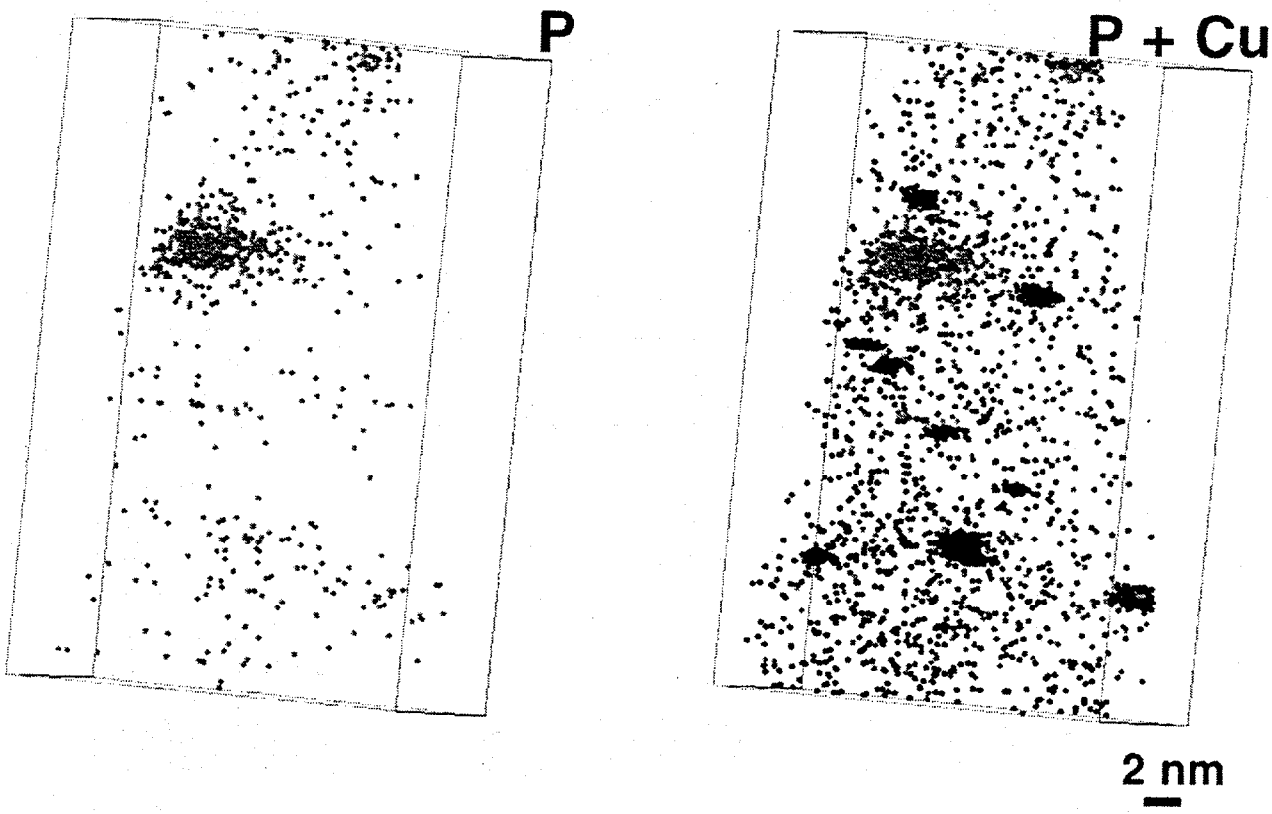

Figure 5. **Atom maps of the phosphorus and copper distribution in weld 73 after neutron irradiation to a fluence of 2×10^{23} n m^{-2} and anneal for 0.5 h at 454°C. A small phosphorus-enriched region is evident.**

Table 3. Number of copper atoms detected in the precipitates and their estimated radii based on the number of copper atoms in the submerged arc weld (weld 73W) for the different heat treatments

Copper-enriched precipitate	Irradiated 2×10^{23} n m^{-2} (E > 1 MeV)		Irradiated + annealed 0.5 h at 454°C		Irradiated + annealed 1 h at 454°C		Irradiated + annealed 168 h at 454°C	
	Ions	r, nm	Ions	r, nm	Ion	r, nm	Ions	r, nm
1	241	0.88	653	1.23	307	0.96	>2287	1.87
2	135	0.73	170	0.79	247	0.89	445	1.08
3	<123	0.71	135	0.73	173	0.79	>35	0.46
4	115	0.69	>122	0.71	163	0.77		
5	84	0.62	75	0.60	122	0.70		
6	43	0.50	75	0.60	120	0.70		
7	36	0.43	60	0.56	115	0.69		
8			34	0.46	89	0.63		
9			>33	0.45	67	0.58		

> Denotes that precipitate was clipped by the analysis volume.

where x_i, y_i, and z_i are the spatial coordinates of each atom; m_i is the mass of each one; and n is the number of atoms in the feature. If all atoms are the same species (i.e., the same mass), this reduces to [11]

$$\bar{x} = \frac{\sum_{i=1}^{n} x_i}{n}, \quad \bar{y} = \frac{\sum_{i=1}^{n} y_i}{n}, \quad \text{and} \quad \bar{z} = \frac{\sum_{i=1}^{n} z_i}{n} \tag{2}$$

The radius of gyration is a measurement of how far from the origin (i.e., the center of mass of the precipitate at $\bar{x}$, $\bar{y}$, $\bar{z}$) the entire mass, m_o, might be concentrated and still give the same moment of inertia, I_x. It is given for the one-dimensional case by

$$l_x = \sqrt{\frac{I_x}{m_o}} = \sqrt{\frac{\sum_{i=1}^{n} m_i (x_i - \bar{x})^2}{\sum_{i=1}^{n} m_i}} = \sqrt{\frac{\sum_{i=1}^{n} (x_i - \bar{x})^2}{n}} \tag{3}$$

where x_i, y_i, and z_i are the spatial coordinates of each atom; m_i is the mass of each one, and n is the number of atoms in the feature. Similarly the radius of gyration, l_g, for the three-dimensional case is given by

$$l_g = \sqrt{\frac{\sum_{i=1}^{n} (x_i - \bar{x})^2 + (y_i - \bar{y})^2 + (z_i - \bar{z})^2}{n}} \tag{4}$$

These estimates assume that the copper distribution defines the extent of the copper-enriched region. These values are summarized for some of the largest copper-enriched precipitates in each treatment in Table 4. A small increase in the radius of gyration was observed with annealing time at 454°C. However, the magnitude of the increase was not statistically significant because of the small number of precipitates encountered and the large variations in their sizes. The variation in the radius of gyration measurements was less than expected from the number of copper atoms in each precipitate. This small variation, coupled with the ramified nature of the precipitates, suggests that the precipitates are far from an equilibrium state during neutron irradiation and the early stages of annealing. It should be noted that these radii of gyration measurements do not take into account the differences in the local magnification between the matrix and the copper-enriched precipitates [10] and are therefore an underestimate of the true size (see below).

Table 4. The radii of gyrations of the precipitates in the submerged arc weld (weld 73W) for the different heat treatments

The order of the data is the same as in Table 3

Copper-enriched precipitate	Irradiated 2×10^{23} n m^{-2} (E > 1 MeV) l_g, nm	Irradiated + annealed 0.5 h at 454°C l_g, nm	Irradiated + annealed 1 h at 454°C l_g, nm	Irradiated + annealed 168 h at 454°C l_g, nm
1	1.00 [1.81]	1.00 [2.06]	1.08 [1.39]	1.47 [2.92]
2	0.96 [1.47]	1.00 [1.47]	0.95 [1.70]	1.79
3	(0.70) [1.28]	1.17 [1.62]	1.10 [1.42]	(0.98)
4	0.78 [1.29]	(0.76) [0.99]	0.97 {1.21]	
5	0.77 [1.26]	0.85 [1.11]	0.88 [1.12]	
6	1.09 [1.47]	0.88 [1.25]	0.92 [1.19]	
7	0.64 [0.80]	0.88 [1.08]	0.88 [1.34]	
8		0.95	0.99 [1.33]	
9		(0.81)	0.98 [1.22]	

( ) Denotes severely clipped precipitate.
[ ] Denotes corrected for local magnification.

The number density of the copper-enriched regions was also estimated from the number of copper-enriched regions in the volume analyzed. The number of particles was taken as either the number of particles whose centers were inside the analyzed volume or the number of particles in the volume that did not intersect the top, left or rear surfaces of the volume. The volume, V, was estimated from the total number of atoms in the volume, N, which was corrected for the detection efficiency of the mass spectrometer, ξ, and the number of atoms per unit volume of the body-centered cubic iron crystal structure (lattice parameter, a_0 = 0.288 nm with 2 atoms per unit cell; that is, $V = N a_0^3/2\xi$. The number densities were estimated to be ~6.4×10^{23} m^{-3} for the neutron-irradiated material and ~6.2, ~6.1, and ~0.5×10^{23} m^{-3} for the materials annealed for 0.5, 1, and 168 h, respectively. As expected because of the coarsening of the copper-enriched regions, the number density decreased slowly over the first hour of annealing and by more than an order of magnitude after 168 h.

The compositional variations of the copper-enriched precipitates were investigated by several different methods. In the first method, the numbers of different types of atoms within a small spherical volume were determined for each copper-enriched region. This measurement yields the average composition of the core of the copper-enriched region. The position of the analysis volume was adjusted to yield the highest local copper concentration within each copper-enriched region. The solute concentration results are summarized in Table 5 for the different treatments. In all cases, the iron, manganese, nickel, and silicon concentrations in the copper-enriched regions were found to be significant. A significant variation in the composition of the individual precipitates was observed. The measured copper levels were significantly different from the equilibrium concentration of the ϵ-Cu phase of ~95% Cu. The correlation between the copper concentration and the concentration of the other solute elements (i.e., iron, manganese, nickel, and silicon) is shown in Fig. 6. All results indicated that high copper levels were correlated with low iron levels. The correlation with the other elements is less defined.

Table 5. Selected volume precipitate compositions in the submerged arc weld (weld 73W) for the different heat treatments

Concentrations are quoted in atomic percent and are listed in order of decreasing number of copper atoms for each material

	% Fe	% Cu	% Ni	% Mn	% Si
Irradiated	45.1	35.9	4.9	10.6	2.8
2×10^{23} n m^{-2}	63.4	24.8	3.0	5.9	1.0
(E > 1 MeV)	45.1	36.8	6.3	9.0	2.1
	52.9	36.1	4.2	3.4	2.5
	78.5	12.7	1.3	2.5	3.8
	61.2	25.4	4.5	6.0	1.5
	54.3	32.8	5.2	5.2	2.6
Irradiated +	36.1	48.1	7.1	4.4	1.1
annealed	59.6	35.1	4.3	1.1	—
0.5 h at 454°C	58.5	36.6	2.4	2.4	—
	49.6	28.5	7.3	9.8	0.8
	72.9	22.0	1.7	—	1.7
	69.8	24.7	1.4	2.7	1.4
	64.8	26.1	3.4	2.3	3.4
	72.3	21.5	1.5	3.1	1.5
	77.8	19.4	2.8	—	—
	32.4	24.3	16.2	18.9	2.7
Irradiated +	50.0	41.1	2.4	3.0	2.4
annealed	56.6	38.6	—	4.8	—
1 h at 454°C	57.7	38.8	1.2	2.4	—
	55.7	36.1	4.1	2.5	1.64
	64.6	27.1	3.1	5.2	—
	68.3	29.3	1.2	1.2	—
	68.6	24.6	3.3	3.3	—
	73.2	17.1	2.4	4.9	1.22
Irradiated +	42.9	51.0	2.76	2.40	0.4
annealed	55.0	30.0	—	15.0	—
168 h at 454°C	48.7	39.0	3.77	8.49	—

Fourteen concentrations not detected.

The difference in the numbers of atoms in these copper-enriched regions was also compared with the number of atoms in similar volumes in the matrix so that the differences in local magnification between the precipitate and matrix regions could be estimated. Variations between ~1.5 and ~4 times the number of atoms in the copper-enriched precipitate and the matrix were measured, and the local magnification was generally higher in precipitates with higher copper contents. These local magnification measurements indicate that the radius-of-gyration results should be increased by from 1.2 to 2 times. The increase in these values due to local magnification differences is in agreement with previous measurements obtained from field ion images on similar thermally aged iron-copper alloys [13]. The corrected values of the radius-of-gyration measurements are included in Table 4. As expected, the

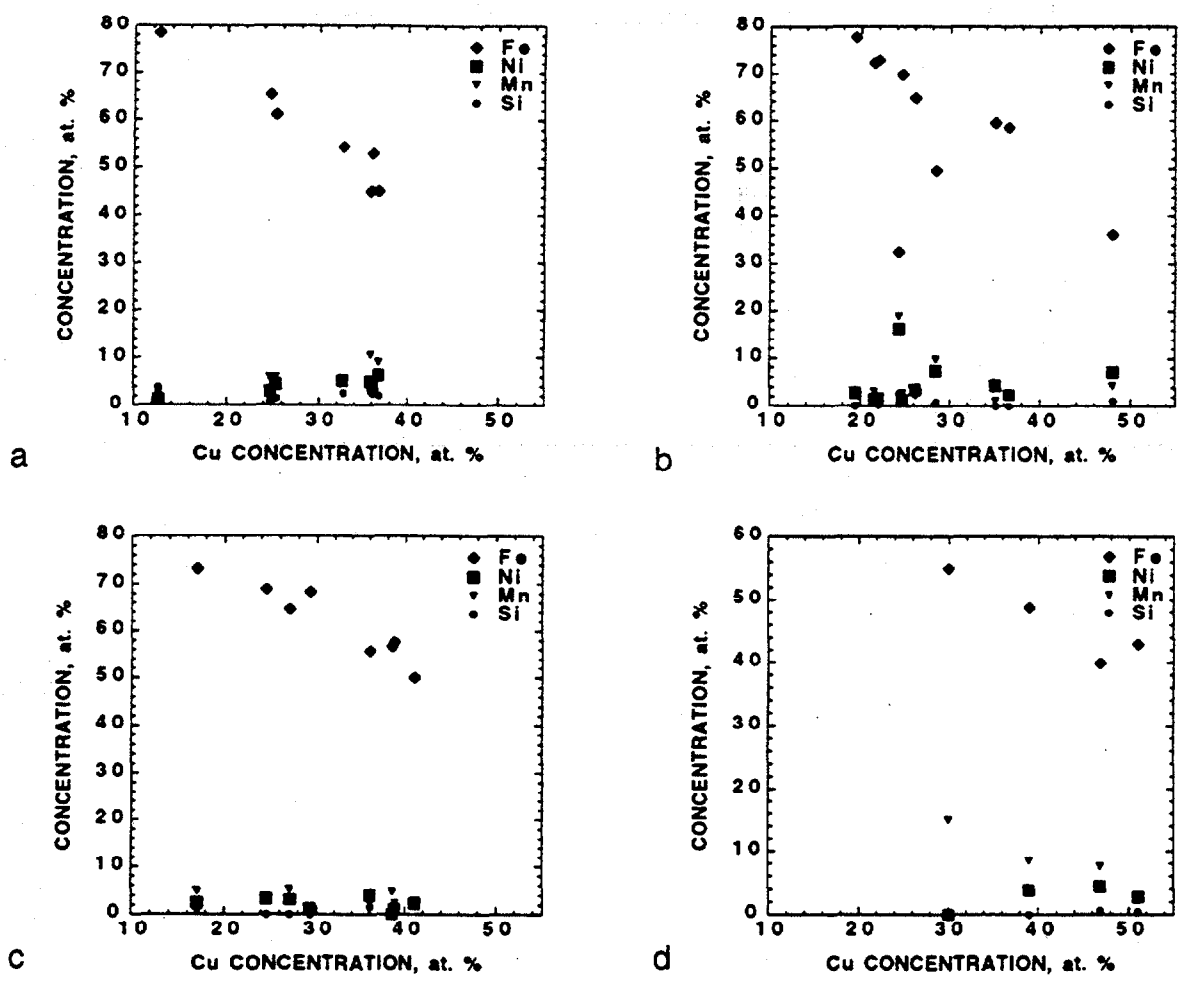

Figure 6. Concentration correlation of the copper-enriched precipitates in weld 73 after (a) neutron irradiation to a fluence of 2×10^{23} n m^{-2}, (b) anneal for 0.5 h, (c) anneal for 1 h, and (d) anneal for 168 h at 454°C.

radius-of-gyration measurements are slightly larger than the estimate based on the number of copper atoms because there are other solutes within the precipitate.

In the second method, the radial concentration profile from the center of the copper-enriched region into the matrix was determined. The center of the copper-enriched region was determined from the distribution of the copper atoms in each copper-enriched region, as described previously. Then the concentrations of the different types of solute atoms that were within spherical shells of equal thickness were determined as a function of distance from the center of the copper-enriched regions. Examples of the results for the different treatments are shown in Figs. 7–10. It should be noted that the ramified or diffuse nature of the copper-enriched regions and small non-uniformities in the shape of each region will influence the sharpness of the interface in these radial composition profiles. The enrichments of copper, manganese, nickel, and silicon and the depletion in iron in the copper-enriched regions are

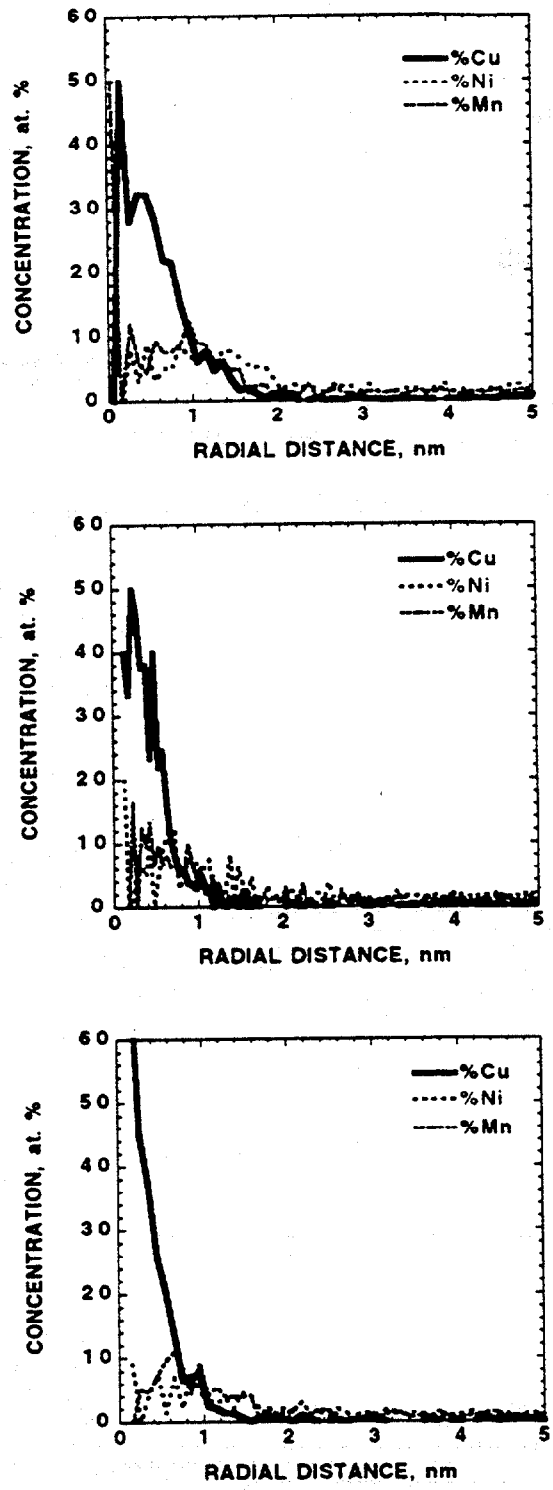

Figure 7. Radial concentration distributions of three copper-enriched precipitates in weld 73 after neutron irradiation to a fluence of 2×10^{23} n m^{-2}.

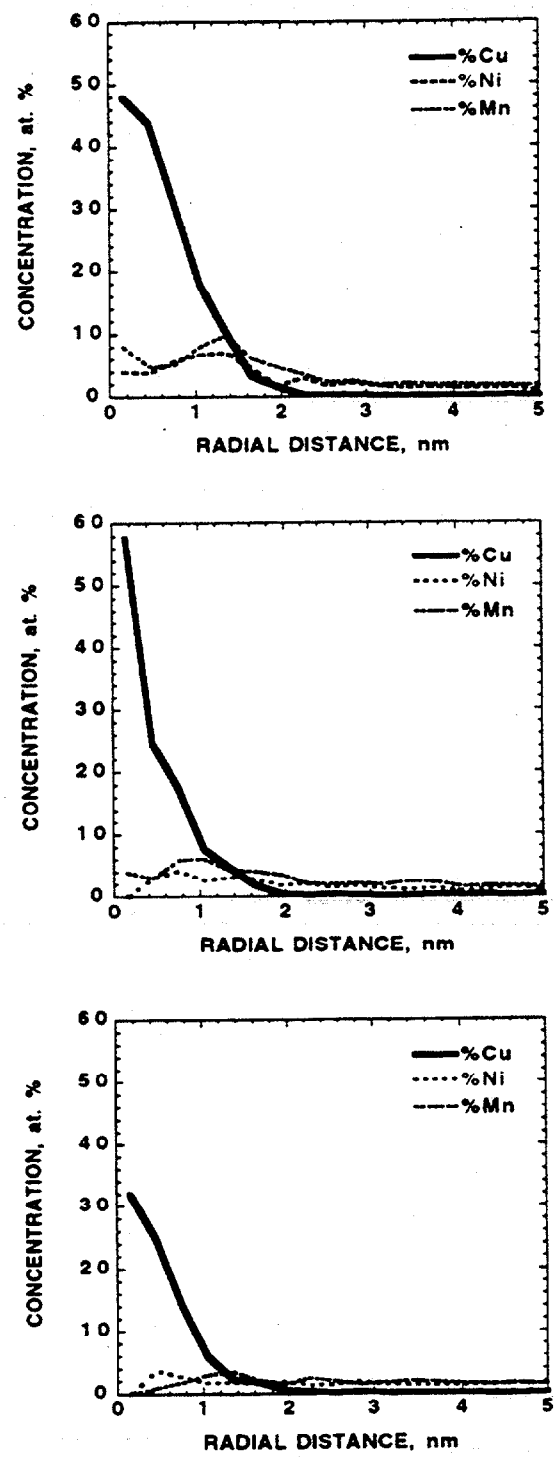

Figure 8. Radial concentration distributions of three copper-enriched precipitates in weld 73 after neutron irradiation to a fluence of 2×10^{23} n m^{-2} and anneal for 0.5 h at 454°C.

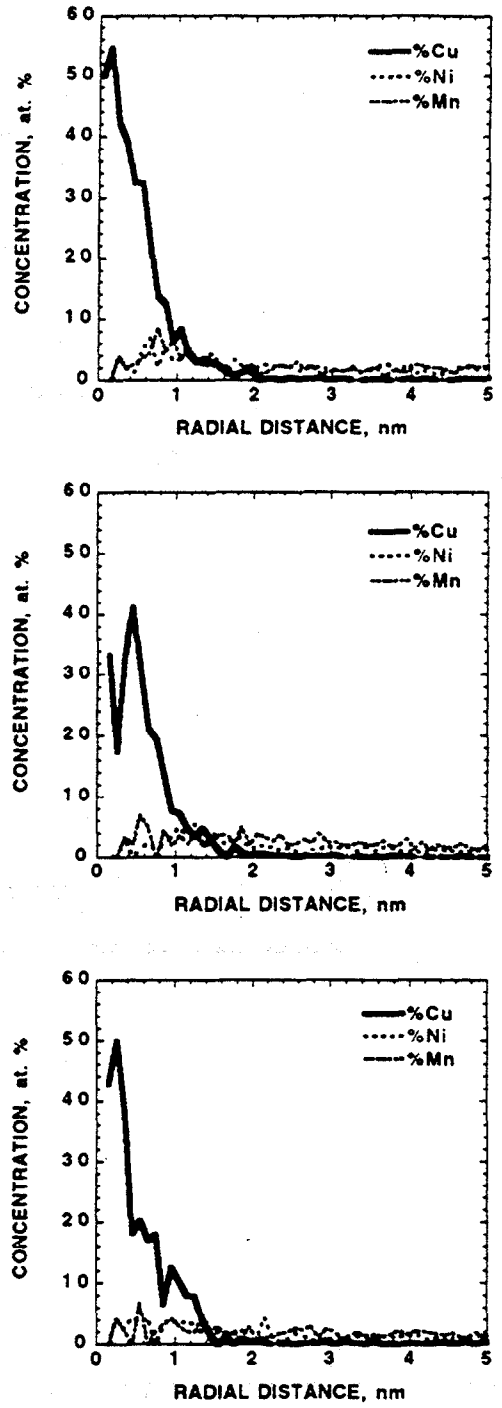

Figure 9. Radial concentration distributions of three copper-enriched precipitates in weld 73 after neutron irradiation to a fluence of 2×10^{23} n m^{-2} and anneal for 1 h at 454°C.

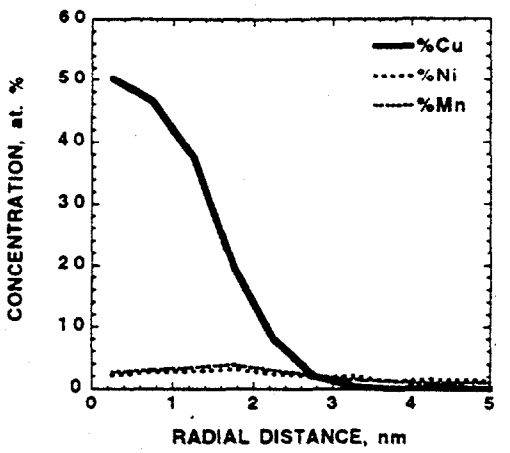

Figure 10. Radial concentration distribution of a copper-enriched precipitate in weld 73 after neutron irradiation to a fluence of 2×10^{23} n m^{-2} and anneal for 168 h at 454°C.

clearly evident. It was also evident that there were some solute enrichments at the precipitate matrix interface. Linear composition profiles through the center of the copper-enriched regions also indicated that the solute enrichments were not always uniformly distributed.

CONCLUSIONS

The size and composition of the copper-enriched precipitates that form during neutron irradiation to a high fluence have been determined by atom probe tomography and atom probe field ion microscopy. As expected, the size was found to increase and the number density of these precipitates to decrease during isothermal annealing at 454°C. Significant levels of iron, nickel, manganese, and silicon were found in the core of the copper-enriched precipitates. Enrichments of nickel and manganese were observed at the precipitate-matrix interface. The distribution of the solute enrichment was not always uniform within the copper-enriched region.

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11. ABSTRACT *(200 words or less)*

A combined atom probe tomography and atom probe field ion microscopy study has been performed on a submerged arc weld irradiated to high fluence in the Heavy-Section Steel Irradiation (HSSI) fifth irradiation series (Weld 73W). The composition of this weld is Fe - 0.27 at. % Cu, 1.58% Mn, 0.57% Ni, 0.34% Mo, 0.27% Cr, 0.58% Si, 0.003% V, 0.45% C, 0.009% P, and 0.009% S. The material was examined after five conditions: after a typical stress relief treatment of 40 h at 607 °C, after neutron irradiation to a fluence of 2×10^{23} n m^{-2} (E > 1 MeV), and after irradiation and isothermal anneals of 0.5, 1, and 168 h at 454 °C. This report describes the matrix composition and the size, composition, and number density of the ultrafine copper-enriched precipitates that formed under neutron irradiation and the change in these parameters with post-irradiation annealing treatments.

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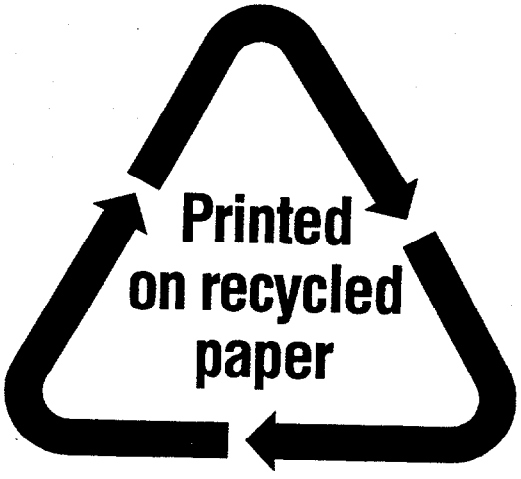

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