AN ATOM-PROBE FIELD-ION MICROSCOPE FOR THE STUDY OF THE INTERACTION OF IMPURITY ATOMS OR ALLOYING ELEMENTS WITH DEFECTS *

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A time-of-flight (TOF) atom-probe field-ion microscope (FIM) designed for the study of defects is described. This atom probe features: (1) a variable magnification internal-image-intensification system; (2) a liquid-helium goniometer stage; (3) a low-energy (<3 keV) gas-ion gun for in-situ irradiations; (4) an ultra-high vacuum (~3 × 10⁻¹⁰ torr) chamber; (5) a high vacuum (~10⁻⁸ Torr) specimen-exchange device; (6) a Chevron ion detector; and (7) an eight-channel digital timer with a ±10 ns resolution for measuring the TOFs. The entire process of applying the evaporation pulse to the specimen, measuring the voltages, and analyzing the TOF data is controlled by a computer. With this system we can record and analyze 600 TOF/min. Results on unirradiated specimens of molybdenum, tungsten, W-25 at% Re, Mo-1.0 at% Ti, Mo-1.0 at% Ti-0.08 at% Zr and a special low swelling stainless steel alloy (LS1A) demonstrate the instrument’s ability to determine quantitatively concentrations at the 5 × 10⁻⁴ at fr level and to determine their spatial distribution with a resolution of a few angstroms.

Un microscope ionique à champ (FIM) et une sonde atomique à temps de vol destinés à l’étude des défauts sont décrits. Cette sonde atomique a les caractéristiques suivantes: 1° un système d’intensification de l’image interne à grossissement variable; 2° une platine goniométrique refroidie à l’hélium liquide; 3° un canon à ions gazeux de faible énergie (3 keV) pour effectuer des irradiations in-situ; 4° un dispositif à ultra-vide (3.10⁻¹⁰ torr); 5° un dispositif de changement de l’échantillon sous vide de 10⁻⁶ torr; 6° un détecteur ionique Chevron et 7° un timer digital à huit canaux avec une résolution de ± 10 nsec de résolution pour mesurer les temps de vol. Le procédé complet d’application du flash d’évaporation à l’échantillon, la mesure des tensions et l’analyse des données de temps de vol est contrôlée par ordinateur. Avec ce système nous pouvons enregistrer et analyser 600 temps de vol par minute. Les résultats sur des échantillons non irradiés de molybdène, de tungstène, d’alliage W-25 at% Re, Mo-1.0 at% Ti, Mo-1.0 at% Ti et 0,08% at Zr et un acier inoxydable spécial à faible gonflement (LS1A) démontrent les possibilités qu’offre cet instrument pour déterminer quantitativement des concentrations de 5 · 10⁻⁴ at fr et pour déterminer leur distribution spatiale avec une résolution de quelques angstroms.

Es wird ein Feldionenmikroskop in Verbindung mit einem speziellen Flugzeitmassenspektrometer beschrieben, das zur Untersuchung von Defekten entwickelt wurde. Diese Atomsonde zeichnet sich aus durch: (1) variable Vergrößerung durch ein internes Bildverstärkungssystem; (2) eine Goniometerbühne für Flüssiheliumtemperaturen; (3) eine Gasionenkanone niedriger Energie (< 3 keV) für in-situ-Bestrahlungen; (4) eine Ultrahochvakuumkammer (~10⁻⁶ Torr); (5) eine Chevron-Ionendetektor; (7) ein digitales Achtkanalzeitwerk mit einer Auflösung von ±10 ns zur Messung der Flugzeiten. Der Gesamtvorgang der Anwendung des Verdampfungspulses auf die Probe, der Spannungsmessung und der Analyse der Flugzeitdaten wird durch einen Rechner kontrolliert. Mit diesem System können 600 Flugzeiten pro Minute aufgezeichnet und analysiert werden. Ergebnisse an unbestrahlten Proben aus Mo, W, W/25 at% Re, Mo/1,0 at% Ti, Mo/1,0 at% Ti/0,08 at% Zr und einer speziellen, niedrig schwellenden rostfreien Stahllegierung beweisen die Fähigkeit des Instruments für eine quantitative Bestimmung von Konzentrationen im Molenbruchbereich von 5 · 10⁻⁴ at fr und deren räumlicher Verteilung mit einer Auflösung von wenigen Å.

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1. Introduction

The invention [1] of the time-of-flight (TOF) atom-probe field-ion microscope (FIM) has provided us with an unique instrument for the study of the interaction of impurity atoms or alloying elements with point, line or...
Fig. 1. A schematic diagram of the TOF atom-probe FIM. Shown at the top of the diagram are the internal elements of the atom probe including the FIM specimen, the internal image intensification system, the focusing lens, the viewing mirror and the Chevron ion detector. The specimen voltage system and the digital timer, shown in the lower portion of the figure, are operated automatically by a Nova 1220 minicomputer which is interfaced to a Tektronix 4010 graphics display terminal and a 4610 hard copy unit so that the data can be plotted on-line during the course of an experiment. With this automated system we can presently record and analyze 600 TOF/min.

The TOF atom-probe FIM consists of a FIM combined with a special TOF mass spectrometer. This spectrometer allows the investigator to identify chemically any atom that appears in a FIM image. Thus, it is now possible both to image the microstructural features of a metal specimen and to measure the mass-to-charge ratios \((m/n)\) of individual atoms from preselected regions of a specimen with a spatial resolution of a few angstroms.

The principles and mode of operation of the atom probe can be understood with the aid of fig. 1. A FIM specimen with a radius of \(\approx 50-500 \, \text{Å}\) is mounted on a liquid-helium cooled goniometer stage and is maintained at a positive potential of \(\approx 5-20 \, \text{kV}\) so that gas atoms (typically helium or neon) are field-ionized over individual atomic sites and are then projected radially outward to produce an image of the atoms on the surface of the metal with the aid of the internal image intensification system (IIFS). In practice the FIM image is observed by means of the front-surfaced glass mirror which is placed at an angle of \(45^\circ\) with respect to the IIFS. The application of a short high-voltage pulse to the FIM specimen results in the field-evaporation of the atoms on the surface of the specimen in the form of positively-charged ions. The ions whose trajectories carry them into the probe hole at the center of the IIFS subsequently travel down the flight tube to a high gain (=10^6) Chevron ion detector. A small fraction of the evaporation pulse is employed to start a high-resolution (\(\pm 10 \, \text{ns}\)) digital timer [2]; the digital timer is stopped by an output pulse from the Chevron ion detector every time an ion arrives at the detector. The TOFs of the ions and the voltage on the specimen are used to determine the \((m/n)\) ratios of the pulse-field evaporated ions. The operation of the atom probe is controlled on-line by a minicomputer which triggers the high-voltage pulse to the tip, reads in the TOF and voltage data, and then calculates the \((m/n)\) values. The minicomputer operates in real time so that the results can be displayed immediately on the graphics terminal as the experiment progresses.

Since the first report of the atom probe [1] other instruments have also been reported in the scientific literature and at recent Field Emission Symposia *.

The major applications of the atom probe, to date, have been to metallurgical problems involving preci-

The atom probe is ideally suited for the study of the interaction of impurity atoms or alloying elements with lattice defects. The basic reason for this is that by controlled pulse-field evaporation of successive layers [9,10] it is possible to examine the bulk of the specimen and to reconstruct in three dimensions the correspondence between specific microstructural features and the local chemical composition. More specifically the atom-probe FIM is capable of imaging lattice defects such as vacancies [11,12], self-interstitial atoms (SIAs) [15], dislocations [16], grain boundaries [17] and voids [18] and it can also chemically identify both substitutional and interstitial alloying elements or impurity atoms. In this paper we briefly describe some of the special features of the atom probe we have constructed (section 2). We also present the results of a number of experiments which demonstrate the instrument’s ability to examine a wide variety of pure metals and metal alloys and to determine the concentration of alloying elements or impurity atoms at the $5 \times 10^{-4}$ at fr level (section 3).

2. Atom-probe field-ion microscope system

2.1. Time-of-flight mass spectrometer

To determine $(m/n)$ values the voltage on the specimen is measured by an analog-to-digital (A/D) converter. The TOF and voltage data is sent to the minicomputer which then calculates the $(m/n)$ ratios employing the equation

$$m/n = 2e(V_{dc} + \alpha V_{pulse})(t - t_0)^2/d^2,$$

(Panitz et al. [19]) where $e$ is the charge on an electron, $V_{dc}$ is the steady-state imaging voltage, $V_{pulse}$ is the evaporation pulse voltage, $\alpha$ is the pulse factor, $d$ is the flight distance, and $(t - t_0)$ is the actual TOF of the ion. The quantity $t$ is the observed TOF and $t_0$ is the total delay time (560 ns). Eq. (1) was derived by equating the kinetic energy of a pulse field-evaporated ion to its potential energy under the assumption that the ion achieves its terminal velocity in a distance that is extremely small compared with $d$. The controls of the dc, pulse and focusing lens power supplies are coupled together so that $V_{pulse}$ and the lens voltage are always a constant fraction of $V_{dc}$; this technique simplifies the calibration procedure and optimizes the resolution of the atom probe [20]. The pulse voltage is typically maintained at $(0.04 - 0.20) V_{dc}$ and the lens is maintained at $(0.3 - 0.5) V_{dc}$. Below $\approx 0.04 V_{dc}$ the pulse behaves erratically and the continuous dc field-evaporation rate becomes comparable to the pulse-field evaporation rate. For values of $V_{pulse} > 0.20 V_{dc}$ the increased spread in the energy of the field-evaporated ions degrades the mass resolution $(\Delta m/m)$ of the spectrometer.

2.2. Internal image-intensification system and focusing lens

The FIM image is observed with the aid of an IIS (see fig. 1) which consists of a high gain ($\approx 10^3$) Galileo 75 mm dia. channel electron multiplier array and a phosphor screen. The entire assembly consisting of the IIS, focusing lens and front-surfaced glass mirror is attached to two ultra-high vacuum metal bellows arranged so that the distance from the tip of the specimen to the front surface of the IIS can be varied continuously from $\approx 12$ to 100 mm; thus providing an areal magnification change of $>64X$.

2.3. Helium-cooled goniometer stage

A liquid-helium cooled goniometer stage has been constructed which has the following features: (1) the specimen can be cooled quickly to cryogenic temperatures to improve the quality and resolution of the FIM image and also to change the diffusivity of point defects; (2) the specimen can be rotated by $\pm 35^\circ$ about two orthogonal axes which intersect at the tip of the specimen so that a selected region of the specimen can be projected over the probe hole for chemical analysis; and (3) the goniometer stage can be translated in three mutually-orthogonal directions to facilitate the alignment of the tip with respect to the probe hole and the low-energy gas ion-gun described in section 2.4. Performance tests showed that the FIM specimen is cooled to 30 K within $\approx 20$ min and to 13 K within 1 h with a liquid-helium consumption rate of $\approx 1.5$ l/h at 13 K. At temperatures greater than $\approx 30$ K the consumption rate drops to $\approx 0.1$ l/h.
2.4. Low-energy gas ion-gun

A low-energy gas ion-gun [21] has been constructed to enable us to irradiate FIM specimens in situ. In operating the ion gun, a gas such as $\text{H}_2$, $\text{He}$, $\text{Ne}$, $\text{Ar}$ or $\text{Xe}$ is continuously bled through a stainless steel cylindrical plasma chamber held at a positive potential. The gas is ionized in the plasma chamber by a magnetically confined 2 mA beam of 30–100 eV electrons emitted from a heated tungsten filament. The ions produced in the plasma chamber are extracted, accelerated and focused onto the FIM specimen which is held at ground potential. The ion gun can produce a 4 cm dia. beam of 100–3000 eV gas ions at a current density of 0.01–10 $\mu$A/cm$^2$ with a gas pressure ranging between $3 \times 10^{-6}$ and $5 \times 10^{-4}$ torr, respectively.

The low-energy ions are used to produce a sea of SIAs by the focused collision replacement (RCS) mechanism. As a result of the RCSs the immobile vacancies remain at the irradiated surface and the SIAs are injected into the tip of the FIM specimen. The vacancies in the surface can be removed by the subsequent field-evaporation of several surface layers. The ion gun is also used to implant gas atoms (e.g. helium or hydrogen) in the tip of the FIM specimen.

2.5. The ultra-high vacuum FIM

The primary vacuum pumps are a titanium sublimation pump (TSP) and a 140 l/s ion pump. The vacuum chamber is initially rough pumped by two sorption pumps used in sequence. A well-trapped 5 cm dia. oil diffusion pump is used to pump the main chamber when the atom-probe FIM contains an imaging gas. At all other times the diffusion pump is valved off to prevent backstreaming of oil into the vacuum system. Ultimate pressures of $3 \times 10^{-10}$ torr are obtained after baking to 150°C for several hours, while under typical operating conditions the pressure is $1 \times 10^{-9}$ torr. The composition of the residual pressure in the vacuum system is measured with a Uthe Technology Inc. (UTI) Model 100C residual gas analyzer.

A specimen-exchange air lock was incorporated into the design of the atom probe to allow the FIM specimen to be replaced without exposing the vacuum system to atmospheric pressure. The pressure in the FIM remains below $10^{-7}$ torr during the exchange and drops to $3 \times 10^{-9}$ torr within 15 min after the exchange port has been closed.

3. Results

In this section we present atom probe results on control specimens of a number of metal alloy systems which we are presently studying. The specific aims of the research program are: (1) the determination of the extent of irradiation-induced segregation to voids in fast-neutron irradiated alloys; (2) the measurement of the number density, size distribution, morphology and chemical composition of irradiation-induced precipitates; and (3) a determination of the extent of redistribution of alloying elements and/or impurity atoms as a result of fast-neutron or heavy-ion irradiation. The objective for all three experiments is to try to understand the data obtained in terms of possible implications on the mechanism(s) responsible for the suppression or enhancement of void formation. A strong effort is also being made to determine the role played by gaseous impurities in the nucleation and stabilization of voids.

Before considering the results we discuss how the data is obtained and displayed. As a FIM specimen is field evaporated the voltage is slowly raised by the investigator to maintain a steady pulse-field evaporation rate. After each pulse the computer reads in the TOF and voltage data, computes the $(m/n)$ values and stores the data on magnetic tape. When sufficient data has been accumulated it is displayed on the graphics display terminal in the desired form. At present the two basic modes of display used are: (1) a histogram of the number of events versus the $(m/n)$ ratio; and (2) a composition profile showing the cumulative number of atoms of a given type, within a specified $(m/n)$ range, as a function of the total number of atoms detected.

A schematic illustration of the physical principle behind a composition profile is illustrated in fig. 2. Fig. 2(a) shows a FIM tip formed from a binary alloy of B (solute) in A (solvent). The tip contains a single precipitate which is rich in B; the solute atoms are denoted by the solid black circles. The magnification is adjusted so that during the course of pulse-field evaporation of the specimen all the atoms (A and B) contained within a cylindrical volume element are chemically analyzed by the atom probe. Fig. 2(b) shows the corresponding composition profile; the number of B atoms versus the cumulative number of A plus B atoms. The cumulative number of atoms is
proportional to depth \( z \), since the specimen is chemically analyzed by dissecting atomic planes sequentially; in a typical experiment the specimen is analyzed to a depth of \( \approx 250 \, \text{Å} \). If the B atoms are distributed uniformly\(^*\) the average slope of the plot in the regions away from the indicated precipitate determines the average composition of the alloy. In the region of the precipitate the average slope of the composition profile yields the precipitate's average composition. The identification of this change in composition as being caused by a precipitate can be confirmed from the microstructural information present in the FIM image. It is also possible to use the composition profiles, in conjunction with the FIM images, to distinguish among: (1) clustering; (2) short-range order (or anti-clustering); (3) composition fluctuations; and (4) pre-precipitation clusters such as Guinier–Preston zones. In this section we use the term “cluster”\(^*\) to indicate any local composition fluctuation which is obviously greater than the composition fluctuations expected in a random solid solution.

\(^*\) We will use the term “uniform” to indicate an absence of any gross segregation effects in the volume of space sampled.

3.1. Molybdenum

The spectrum of a pure molybdenum specimen is shown in fig. 3 for \( m/n \) ranging from 0 to 100 amu. Molybdenum field evaporates in three charge states (\( \text{Mo}^{2+} \), \( \text{Mo}^{3+} \) and \( \text{Mo}^{4+} \); \( \text{Mo}^{3+} \) is the most abundant state and \( \text{Mo}^{4+} \) is the least abundant one. Note the presence of the two relatively small peaks of \( \text{H}^{+} \) and \( \text{He}^{+} \) caused by residual hydrogen and helium in the FIM. Fig. 4

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* We will use the term “uniform” to indicate an absence of any gross segregation effects in the volume of space sampled.
shows the $\text{Mo}^{2+}$ portion of the spectrum and it is evident that the seven stable isotopes of Mo ($^{92}\text{Mo}$, $^{94}\text{Mo}$, $^{95}\text{Mo}$, $^{96}\text{Mo}$, $^{97}\text{Mo}$, $^{98}\text{Mo}$ and $^{100}\text{Mo}$) are resolved; in addition we have obtained isotopic abundances which agreed very well with the handbook values [21,22].

3.2. Tungsten and tungsten –25 at% rhenium

The addition of Re to W strongly suppresses swelling (void formation) in fast-neutron irradiated W(Re) alloys [23]. It is our objective to determine how the Re is redistributed as a result of the fast-neutron irradiation treatment and to study the irradiation-produced precipitates in this alloy. The results on W and W–25 at% Re will serve as a control for some in-progress experiments.

The peaks associated with the five stable isotopes of W ($^{180}\text{W}$, $^{182}\text{W}$, $^{183}\text{W}$, $^{184}\text{W}$, and $^{186}\text{W}$) are readily distinguished in the $\text{W}^{3+}$ spectrum shown in fig. 5(a). The spectrum for the W–25 at% Re alloy is shown in fig. 5(b). The peaks due to the two isotopes of Re ($^{185}\text{Re}$ and $^{187}\text{Re}$) are clearly resolved from the immediately adjacent W isotopes. For both pure W and the W(Re) alloy the isotopic abundances are in good agreement with the handbook values [21,22]. The average composition of the Re in the W(Re) alloy was determined from the composition profiles shown in fig. 6. The average composition for the $^{185}\text{Re}$ isotope is 10.7 at% and for the $^{187}\text{Re}$ isotope it is 14.7 at%; hence the overall Re composition is 25.4 at%. The good agreement between the measured and nominal Re concentration and the FIM image indicates that all the Re atoms are in solid solution. A second conclusion is that there was no evidence for any pre-precipitation. A third conclusion is that the composition profiles exhibit composition fluctuations about the average Re isotope compositions (see fig. 6). To date we have been unable to conclude whether or not there is any evidence for short-range order (anti-clustering) obtainable from the composition profiles.

3.3. Molybdenum (titanium) and molybdenum (titanium and zirconium) alloys

Both Mo–1.0 at% and Mo–1.0 at% Ti–0.08 at% Zr (TZM) exhibit an enhanced swelling behavior when
Fig. 6. (a) A composition profile for the $^{187}$Re isotope in the W–25 at% Re alloy. The number of events in the range 62.3–62.6 amu are plotted as a function of the cumulative number of events in the W–Re$^{3+}$ spectrum from 59.9 to 65 amu. (b) A composition profile for the $^{185}$Re isotope in the W–25 at% Re alloy. The number of events in the range 61.6–62.0 amu is plotted as a function of the cumulative number of events in the W–Re$^{3+}$ spectrum from 59.9 to 65 amu.

compared to molybdenum specimens irradiated under identical conditions to the same fast-neutron dose [23,24]. At present we are engaged in a study to determine the role(s) played by the Ti in Mo–1.0 at% Ti and the Ti and Zr in TZM in causing this swelling enhancement. It is interesting to note, by contrast, that Johnston et al. [25] have found Ti and Zr additions to be effective swelling inhibitors in a series of model Fe–Cr–Ni alloys.

The Ti$^{3+}$ spectrum for the as-received Mo–1.0 at% Ti alloy is shown in fig. 7; the five isotopes (46Ti, 47Ti, 48Ti, 49Ti and 50Ti) are very well resolved. The agreement between the measured and the handbook isotopic abundances was found to be reasonable in view of the fact that the sample size only involved 30 Ti$^{3+}$ ions for all five isotopes. The five Ti$^{3+}$ isotopes present a very decisive indicator of Ti which allows us to distinguish it from O; oxygen has only three isotopes (16O, 17O and 18O) with 16O having a natural abundance of 99.8 at%. The concentration of Ti was determined to be 0.28 at%; this represents 28% of the expected 1 at% Ti concentration. There may also be additional Ti atoms which field evaporate as Ti$^{2+}$ and they would, therefore, be superimposed on the Mo$^{4+}$ portion of the spectrum. However, it is emphasized that Chang and Perlmutter [26] have shown that Mo$_2$C, TiC and ZrC precipitates can form in both Mo(Ti) and TZM. Thus, the remaining 72% of the Ti may alternatively be tied up in the form of TiC precipitates which exist at a number density that is below the limit of detection of the FIM. The Ti observed was found to be uniformly distributed throughout the alloy. This is significant because a comparison of the present results with the results on the irradiated Mo(Ti) specimens should allow us to comment in detail on the changes produced in the spatial distribution of Ti atoms in the alloy as a result of the fast-neutron irradiations.

Fig. 8 shows a semi-logarithmic plot of the (m/n) histogram of TZM between 29 and 35.5 amu for Zr$^{3+}$ and Mo$^{3+}$. The isotopes of Zr are $^{90}$Zr, $^{91}$Zr, $^{92}$Zr, $^{94}$Zr and $^{96}$Zr; hence there is an almost exact overlap with $^{92}$Mo, $^{94}$Mo and $^{96}$Mo. The $^{90}$Zr and $^{94}$Zr isotopes have isotopic abundances of 51.46 and 11.23 at%, respectively; hence they constitute 62.69 at% of the five Zr isotopes. We also observed $^{92}$Zr$^{2+}$ at 45 amu; hence the total Zr concentration
is 0.13 at% (based on the isotopic abundance of $^{90}\text{Zr}$) which is in reasonable agreement with the nominal Zr concentration.

The composition profiles of Ti and Zr in TZM are shown in fig. 9; each step in the ordinate scale corresponds to single Ti or Zr atom. The Ti atoms observed were distributed uniformly throughout the specimen; from the slope of the composition profile the Ti concentration is 0.26 at%. This value represents 26% of the nominal Ti concentration in TZM. The comment made with respect to the difference between the measured and the nominal Ti concentration in the Mo(Ti) alloy also applies to TZM. All the Zr atoms detected were contained in a single cluster; this Zr cluster was detected just before the FIM tip failed and it may have contributed to the failure of the tip. These control experiments on TZM demonstrated that the Ti and Zr alloying elements in the <0.5 at% range could be easily detected and that the techniques developed to date should work reasonably well for the neutron-irradiated specimens.

### 3.4. Low swelling stainless steel alloy (LS1A)

A low swelling stainless steel alloy (LS1A) developed at the Oak Ridge National Laboratory (ORNL) by Bloom et al. [27] has been analyzed. To date we have restricted our work to unirradiated specimens of this alloy because of the high radioactivity of the neutron-irradiated specimens. The alloy LS1A contains 2.06 at% Si and 0.16 at% Ti as swelling inhibitors. It is the purpose of the present study to determine the role played by these swelling inhibitors.

All the alloying elements (Fe, Cr, Ni, Mn, Mo, C, Si and Ti) in LS1A were readily identified and the concentrations measured by the atom-probe techni-
Fig. 10. The 24–34 amu portion of the LSI A spectrum. The main alloying elements (Fe, Ni and Cr) as well as one minor alloying addition (Mn) are seen in this part of the spectrum. This spectrum was recorded at a pressure of $1 \times 10^{-9}$ torr and a specimen temperature of 50 K. The voltage range was from $V_{dc} = 16600 - 19900$ V and the calibration parameters were $a = 1.482$, $t_0 = 0.56 \mu$s and $d = 2.227$ m.

The isotopes $^{50}$Cr$^{2+}$, $^{52}$Cr$^{2+}$, $^{54}$Fe$^{2+}$, $^{55}$Mn$^{2+}$, $^{56}$Fe$^{2+}$, $^{58}$Ni$^{2+}$ and $^{60}$Ni$^{2+}$ can be clearly identified in the 24–34 amu portion of the spectrum. In addition, an analysis of the spatial distribution of each alloying element revealed the existence of a cluster consisting of three nickel atoms, three silicon atoms, two carbon atoms, two titanium atoms, two molybdenum atoms, one manganese atom and one iron atom. It is highly improbable for such a cluster to be the result of a simple statistical fluctuation.

4. Conclusions

In this paper we have described a computer controlled atom-probe FIM which is suited for the study of the interaction of impurity atoms or alloying elements with defects. Detailed results were presented on unirradiated specimens of Mo, W, W–25 at% Re, Mo–1.0 at% Ti, Mo–1.0 at% Ti–0.08 at% Zr and a low swelling stainless steel alloy (LSI A) which demonstrate that both the concentration and the spatial distribution (on an angstrom scale) of each alloying element can be determined quantitatively at levels as low as $5 \times 10^{-4}$ at fr. We are presently applying this technique to fast-neutron irradiated specimens of the above alloys in an attempt to solve the problems enumerated at the beginning of section 3.

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References


Discussion

R. Bullough: How do you know the images in Mo are voids. Have you identified solute segregation on the void surfaces?

D.N. Seidman: First, the micrograph I showed of voids in a neutron-irradiated molybdenum specimen only involved a field-ion microscope examination [S.S. Brenner and D.N. Seidman, Rad. Eff. 24 (1975) 73] and not an atom-probe field-ion microscope examination. The atom-probe study to detect segregation on the void surfaces, which we describe in our paper, is presently in progress. Secondly, we know that the observed images are caused by voids for the following reasons: (1) the contrast effects observed do not occur in unirradiated molybdenum specimens; (2) an electron microscope examination of another portion of the specimen we examined by the FIM technique revealed a lower number density than the void number density we measured; and (3) the range of observed contrast effects we detected for voids are consistent with some electrolytic tank experiments, reported in the literature [P.J. Birdseye, D.A. Smith and G. D.W. Smith, J. Phys. D.7 (1974) 1642], performed to model possible contrast effects from voids.

L.K. Mansur: When a defect is observed in the FIM, say an interstitial, presumably the atoms surrounding it are in distorted positions. What relation can be deduced to the local field of the defect in the bulk? What accuracy would you ascribe to such a determination?

D.N. Seidman: A few years ago we examined this problem in detail [D.N. Seidman and K.H. Lie, Acta Met. 20 (1972) 1045] under the assumption that the configuration of the self-interstitial atom (SIA) retained its identity (i.e. no atomic relaxations were allowed) near the surface of the FIM tip. If this assumption is valid then it is possible to calculate contrast patterns for the different SIA configurations; we presented calculated contrast patterns for the (110) dumbbell and the (111) split crowdion in a bcc lattice in this paper. Experimental evidence has also been presented to show that the SIA does, indeed, give rise to a contrast pattern. Unfortunately, it has not been possible to use the contrast patterns to identify the configuration of the SIA atom. I suspect that the problem lies with the fact that the SIA near the surface of the FIM tip does not retain its bulk identity.

A.D. Brailsford: Because of the large curvature near the periphery of an exposed void, the surface electric stress must be very large. Have you considered the effect of such a stress on the possible solute segregation in its vicinity?
D.N. Seidman: The curvature near the periphery of an exposed void is probably no greater than the "curvature" associated with a single atom in a high Miller indexed plane like the \{111\}, \{332\} or \{343\} planes of a bcc lattice. Thus, I would not expect the local electric field near the periphery of the exposed void to be abnormally high. The effect of the electric field on the diffusion coefficient of an adatom has been studied by several investigators [e.g. see D.W. Basset and M.J. Parsley, Brit. J. Appl. Phys. (J. Phys. D) 2 (1969) 13; W.R. Graham and G. Ehrlich, Surface Sci. 45 (1974) 530; and T.T. Tsong and G. Kellog, Phys. Rev. B12 (1975) 1343]. The general conclusion is that at the normal cryogenic temperatures (<100 K) employed in field-ion microscopy the imaging electric field cannot cause a field-induced motion of adatoms. Thus, the imaging electric field probably doesn't change the distribution of solute atoms which segregate to a void.