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Positron Annihilation Study of Irradiated Iron Alloys

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Positron annihilation lifetime measurements and angular correlation measurements have been performed for pure Fe and Fe alloys to investigate the fundamental behaviors of radiation induced defects and alloying elements. It was found that Cu atoms interact with vacancies and form complexes of Cu and vacancies, which can be considered to be radiation enhanced precipitation of Cu atoms in Fe matrix. Angular correlation measurements of neutron irradiated Fe-Cr-Ni alloys and fatigued 316 ss showed that some vacancy clustering occurred. Calculation of positron annihilation lifetime was performed for a vacancy in Fe on the basis of local density approximation (LDA) with and without lattice relaxation and showed that lattice relaxation considerably reduced the lifetime, resulting in the reasonable fitting with the experimental value.

KEYWORDS: positron lifetime, angular correlation, Fe-Cu, 316ss, n-irradiation

1 Introduction

In order to understand the radiation effect on the materials used in high temperature and high dose environment, such as fission and fusion reactor materials, it is well recognized that fundamental behaviour of radiation induced defects must be clearly investigated. Positron annihilation study has become a very powerful technique for this purpose last decades. On the other hand, computer simulation of the lattice defects and positron lifetime calculation based on the local density approximation and the superposition of atomic wave function have been developed, having made it possible to compare the experimental result and calculation. Interatomic potentials required for the lattice relaxation have been recently developed, such as EAM (embedded atom method) type potential, Finnis Sinclair potential and others. These potentials have rather simple functional form and have made it possible to calculate defect structures in a considerably large model crystal without a big effect of boundary condition. In this report the experimental results for irradiated Fe alloys and some simulation calculations will be presented.

2 Experimental

Fe alloys, mainly Fe-Cu, Fe-Cu-C, Fe-Cr-Ni alloys were prepared in high-purity hydrogen atmosphere, namely, floating zone levelling in hydrogen gas, by using high-purity starting materials. Size of specimens for positron annihilation lifetime measurement was $8 \times 8 \times$ 0.2mmt and $10 \times 15 \times$ 0.2mmt for angular correlation measurement. Neutron irradiation was performed to a dose of 1×10^{17} n/cm^2 in JMTR (Hydraulic Rabbit). Electron irradiation was carried out to 4×10^{18} e/cm^2 at 77K with KURRI LINAC. Positron annihilation lifetime measurement was performed by using a conventional fast-slow coincidence circuit (ORTEC system) with phototubes (Hamamatsu H3378) and BaF2 scintillation counters. Resolution time is about 210 psec. Angular correlation curve was obtained by using the conventional coincidence measurement system with the angular resolution 0.63mrad (FWHM). Cu^{64} (about 1 Ci) was used as a positron source. Analysis of positron annihilation lifetime spectra was performed by Resolution and Positronfit programs. Furthermore, Contin-Pals2 program was also used for some special cases where conventional analysis is rather difficult because of too many positron trapping sites.

3 Results and Discussion

3.1 Neutron irradiated Fe-Cu alloy

Positron annihilation lifetime measurement was performed for the isochronal annealing process of the neutron irradiated Fe-0.22%Cu alloy (JMTR, 1×10^{17} n/cm^2, 100 °C) and the result is shown in Fig.1. The long lifetime component of about 300 psec corresponding to microvoids was observed in the as-irradiated state, but the lifetime decreased by the isochronal annealing thereafter. Microvoids are considered to be formed at cascade sites generated by neutron irradiation. By the isochronal annealing microvoids usually grow in size by absorbing mono vacancies emitted from unstable smaller microvoids. The result in Fig. 1 is, therefore, very unusual and is considered to be due to the interaction between microvoids and Cu atoms. From the phase diagram of Fe-Cu alloy it is clear that solubility limit of Cu atoms in Fe matrix is very small and Cu atoms have a tendency of precipitate formation (1). It is reasonably considered that surface of a microvoid is a suitable site for this precipitation. Since a Cu atom is an oversized atom in Fe matrix (about 17% over in volume, Cu atoms migrate by vacancy mechanism in Fe matrix. Then, Cu atoms arrive at microvoid surfaces and replace Fe atoms at the surface, namely, forming Cu coating at the surface. These coated microvoids are considered to be more stable than uncoated microvoids. But it is not enough to explain the decrease of lifetime with increasing annealing temperature in Fig. 1.

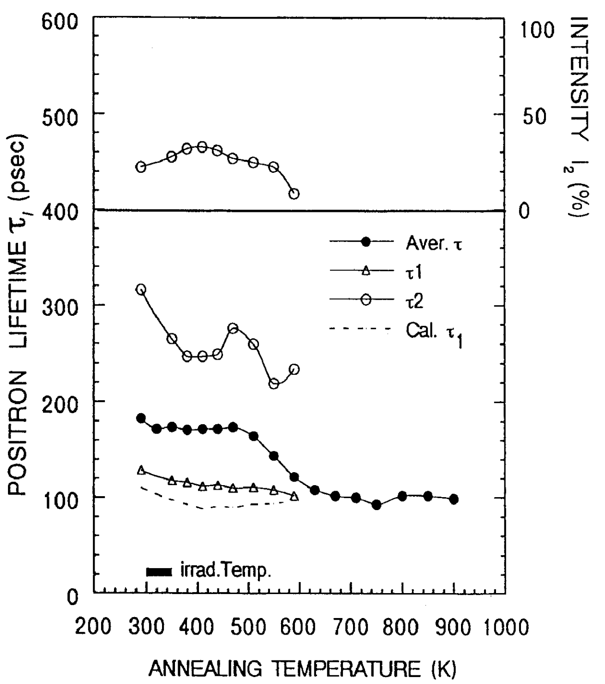

Fig.1 Isochronal annealing result of neutron-irradiated Fe-Cu alloy

One possibility is as follows. Microvoids have a size

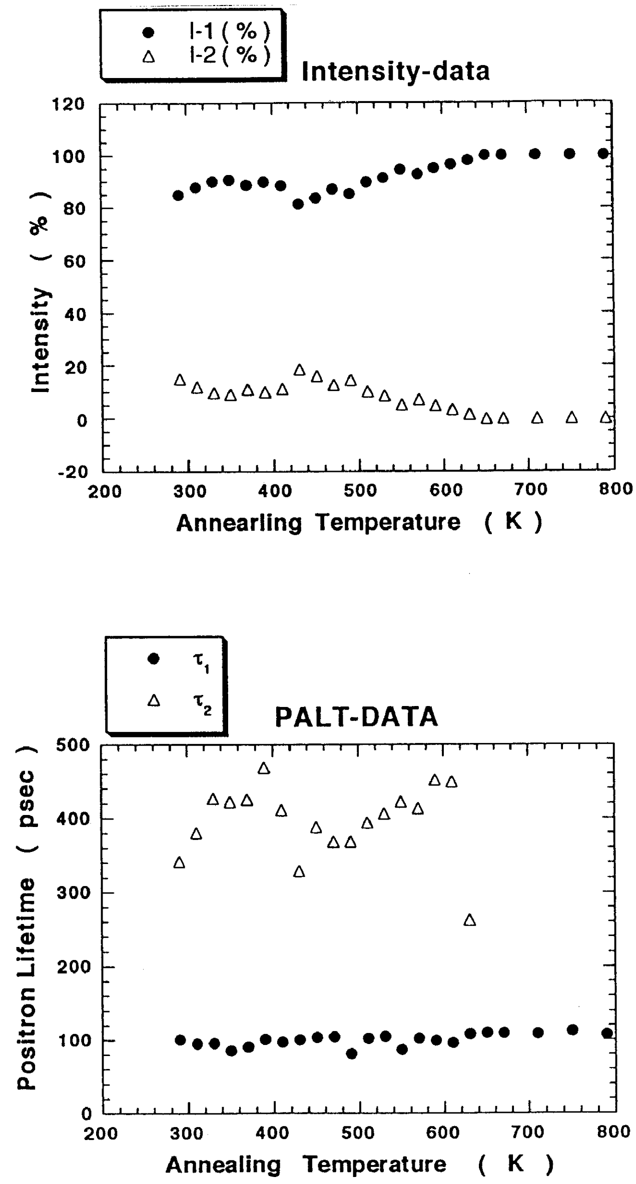

Fig.2 Isochronal annealing result of Cu ion irradiated Fe

distribution and probability of Cu coating on the microvoid surface is higher in smaller microvoids and lower in larger microvoids, because specific sink efficiency of a microvoid for arriving Cu atoms is $4\pi r$ (r is a microvoid radius) and the surface area is $4\pi r^2$. Smaller microvoids well coated have rather higher stability than larger microvoids less coated. This can explain the decreasing tendency of lifetime with increasing annealing temperature because larger microvoids less coated by Cu atoms will be evaporated or shrink to a certain size where the number ratio of Cu atoms over vacancies is large enough for the microvoid to be stable even at higher temperatures and smaller microvoids well coated will remain. But, still it is not enough to explain the behaviour of the second intensity I_2, which rather increases. This suggests that Cu precipitates are formed in the matrix probably with the aid of vacancies emitted from unstable microvoids and act as positron trapping centers, where positron lifetime is shorter than that at microviods. Above this temperature range lifetime again increased and decreased, which suggests that the process of thermal emission and condensation of vacancies with Cu atoms was repeated up to the final recovery stage above 600 K.

3.2 Heavy ion irradiation

To compare the result mentioned above (Fig.1) the same experiment was performed for the 4MeV Cu ion irradiated Fe (at RT, 0.8 dpa) and is shown in Fig.2. In this case damaged region is only 1.2 μm from the surface and final Cu content is 0.3 % in range average. Some of the positrons are trapped at damaged region, but others go into the deeper region where damage does not exist. This is the reason that the second intensity I_2 is so low (about 15 %). The long lifetime corresponding to microvoids is larger here than neutron irradiated case, probably because irradiation dose is higher here (0.8 dpa) than neutron case (about 10^{-4} dpa). The same tendency is seen in the annealing process, namely, decrease and increase of long lifetime is again observed. Essentially, this is the same phenomenon as that mentioned above, that is, the emission of vacancies from microvoids and coprecipitation of vacancies and Cu atoms during annealing process.

3.3 Fundamental property of a vacancy in Fe and Fe-Cu

Recent development of calculation method of the positron lifetime in metals has made it possible to compare the experimental results with calculated results. In this procedure it is very important to take into account the lattice relaxation around a vacancy. Recently a big progress has been achieved in the interatomic potential used for the relaxation calculation, e.g., EAM potential (embedded atom method) and others, which are based on the density functional theorem and have many body character. Relatively simple forms of these potentials have made it possible to relax the reasonable large model lattice within short enough time. Here the potential for Fe obtained by Finnis and Sinclair (3) was used. For the relaxed structure of a vacancy thus obtained the calculation of a positron wave function was made by the method developed by Puska and Nieminen (4), where electron wave functions of each atom (Herman and Skillman (5)) are superimposed and the so-called local density approximation is used to determinethe correlation energy for a positron and an electron. The Schrödinger equation was solved by the method by Kimball and Shortley (6) These are shown bellow:

$$\left[-\nabla^2 + V(\boldsymbol{r})\right] \psi_+(\boldsymbol{r}) = E_+ \psi_+(\boldsymbol{r}) \tag{1}$$

$$V(\boldsymbol{r}) = V_C(\boldsymbol{r}) + V_{corr}[n_-(\boldsymbol{r})] \tag{2}$$

$$V_C(\boldsymbol{r}) = \sum_i \left[\frac{2Z}{r_i} - \frac{2}{r_i}\int_0^{r_i} \rho(t)dt - 2\int_{r_i}^{\infty} \frac{\rho(t)}{t}dt\right] \tag{3}$$

$$V_{corr}(r_s) = \begin{cases} -\frac{1.56}{\sqrt{r_s}} + (0.051\ln r_s - 0.081)\ln r_s + 1.14 & \text{for } r_s \le 0.302, \\ -0.92305 - \frac{0.05459}{r_s^2} & \text{for } 0.302 \le r_s \le 0.56, \\ -\frac{13.15111}{(r_s+2.5)^2} + \frac{2.8655}{r_s+2.5} - 0.6298 & \text{for } 0.56 \le r_s \le 8.0, \\ -179856.2768n^2 + 186.4207n - 0.524 & \text{for } 8.0 \le r_s < \infty \end{cases} \tag{4}$$

$$r_s = \left[\frac{3}{4\pi n_-(\boldsymbol{r})}\right]^{1/3} \tag{5}$$

$$\lambda = \pi r_0{}^2 c \int d\boldsymbol{r} n_+(\boldsymbol{r})[n_v(\boldsymbol{r})\Gamma_v(n_v(\boldsymbol{r})) + n_c(\boldsymbol{r})\Gamma_c + n_d(\boldsymbol{r})\Gamma_d] \tag{6}$$

$$n_+(\boldsymbol{r}) = |\psi_+(\boldsymbol{r})|^2 \tag{7}$$

$$\Gamma_v(n_v) = \left[1 + \frac{r_{s,v}^3 + 10}{6}\right] \tag{8}$$

$$r_{s,v} = \left[\frac{3}{4\pi n_v}\right]^{1/3} \tag{9}$$

$$\tau = \frac{1}{\lambda} \tag{10}$$

,where $V(\boldsymbol{r})$ in Schrödinger equation is the potential for a positron. $V_C(\boldsymbol{r})$ is the Coulomb potential, $V_{corr}(\boldsymbol{r})$ is the correlation energy for a positron and an electron. λ is the annihilation rate, τ is the positron lifetime. In Fig.3 the calculated wave function of a positron trapped at a relaxed vacancy site in Fe is shown. The corresponding positron lifetime is 181 psec which is shorter than the positron lifetime in an unrelaxed vacancy 187 psec. In this case the enhancement factor for d-electrons d was adjusted as 2.20 so that the positron lifetime in the Fe matrix becomes 110 psec. The lifetime calculation was also made for the vacancy relaxed by the tight binding method presented by Masuda and the shorter lifetime 173 psec was obtained. The amount of the relaxation of the nearest neighbour lattice point around a vacancy was $-0.02765a$ (a: lattice constant of Fe) in Finnis-Sinclair potential and $-0.0368a$ in the Masuda's tight binding method, respectively. Here, $-$ means an inward shift.

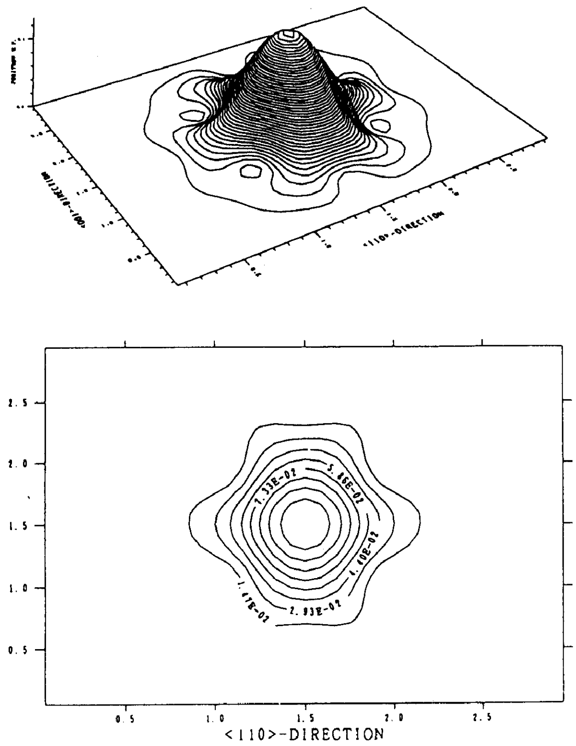

Fig.3 Calculated wave function of a positron trapped at a relaxed vacancy in Fe

The positron annihilation lifetime obtained in the experiment is 170 $\sim$ 175 psec (7). It is then considered that the lattice relaxation is very important in this type of calculation. To improve the calculation more realistic electron structure and the effect of a positron on the relaxed structure of a vacancy must be taken into account.

3.4 Angular correlation measurement of fatigued stainless steel

Angular correlation measurement has been proved to be very effective for the detection of positronium, e.g., in neutron irradiated Nb, V and stainless steels (Fe-Cr-Ni)(2), where the very narrow angular correlation curves were observed. In Fig.4 the angular correlation curve of Fe-15Cr-16Ni neutron irradiated (JOYO, 2.2 $\times$ 10^{22} n/cm^2, 500 °C) is shown. In the present case the same type of measurement was performed for the fatigued 316 stainless steel to detect the vacancy clusters formed by cyclic motion of dislocations. In Fig.5 the angular correlation curve of 316 ss fatigued under the condition N_f = 1.14 $\times$ 105, $\sigma_a = 0.35\sigma_b$. FWHM of the curve is about 10 mrad, which is shorter than that of solution annealed specimen, i.e., 12.5 mrad. But no narrow component was observed, which means vacancies formed through cutting of cyclically moving dislocations do not form prominent microvoids. This is due to small mobility of vacancies at room temperature in this alloy, namely, the migration energy is 1.36 eV. In near future high tem-

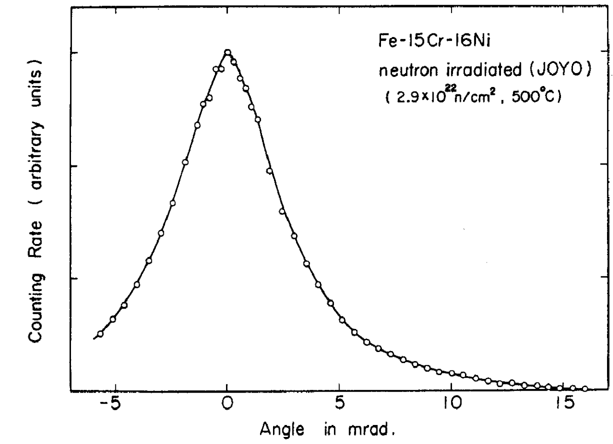

Fig.4 Angular correlation curve of Fe-Cr-Ni alloy irradiated by neutrons in JOYO

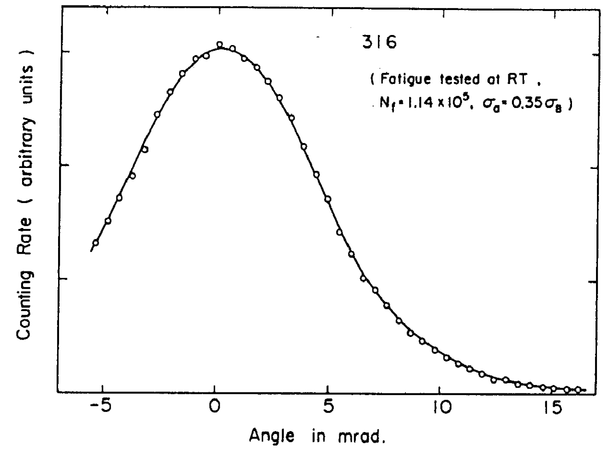

Fig.5 Angular correlation curve of fatigued 316 stainless steel

perature fatigue test will be performed, where clustering of vacancies will be more prominent.

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References

1) F. Hori, Y. Aono, M. Takenaka and E. Kuramoto: *Scripta Met.* 29 (1993) 243.
2) E. Kuramoto, N. Kikuchi, D. Irisawa, M. Takenaka and M. Hasegawa: *Positron Annihilation*, ed. L. Dorikens–Vanpraet, M. Dirikens and D. Segers, World Scientific, (1989) 446.
3) M. W. Finnis and J. E. Sinclair: *Phil. Mag. A*, 501 (1984) 45 (Erratum; *Phil. Mag. A*, 531 (1986) 161).
4) M. J. Puska and R. M. Nieminen: *J. Phys. F: Met. Phys.* 13 (1983) 333.
5) F. Herman and S. Skillman: *Atomic Structure Calcu-*

lations, Prentice Hall, Inc. (1963).
6) G. E. Kimball and G. H. Shortley: *Phys. Rev.* 45 (1934) 815.
7) E. Kuramoto, S. Nagano, K. Nishi, Y. Aono and M. Takenaka: *Materials Science Forum*, 105–110 (1992) 1125.