Positron Annihilation Study of Electron-Irradiated FeCr Alloys

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1. Introduction

Reduced-activation ferritic / martensitic [RAFM] steel is a primary candidate alloy as Gen-IV and fusion reactor materials, because of its many attractive thermophysical properties such as a higher swelling resistance, higher thermal conductivity, lower thermal expansion and better liquid-metal compatibility when compared with austenitic stainless steels. It is known that the amount of Cr in an alloy determines the properties of that material. Especially, high-chromium (9-12%Cr) steels are appealing due to their excellent irradiation resistance to a void swelling [1,2].

Positron annihilation lifetime spectroscopy(PALS) has been applied to investigate the production of vacancy-type defects for electron-irradiated pure Fe, Fe-5Cr, Fe-9Cr, Fe-15Cr alloys.

2. Experimental and Methods

2.1 Experimental and Sample Examined

The PAL measurements were made using a conventional fast-fast coincidence system. The instrumental time resolution of the system was 250 ps of the full width at a half maximum (FWHM) The source correction was made using a well-annealed iron sample, which has a known positron lifetime of 106 ps. We applied the PALSfit program from the RISØ laboratory to analyze the measured spectra [3]. In performing the PALS analysis with the PALSFit package, we used two Gaussian resolution functions to extract the positron lifetime information from the lifetime distributions.

We have examined pure Fe, Fe-5Cr, Fe-9Cr, Fe-15Cr supplied by Hankook Vacuum Metallurgy Co., Korea. All the samples were encapsulated in a quartz tube and annealed in a vacuum at 800 °C for six hours and then slowly cooled down to room temperature in a furnace over several hours. The size of the samples was $10x10x0.4mm^3$. They were irradiated with 2MeV electrons in the first accelerator facility of JAEA Takasaki at 77 K with $1x10^{18}$ e/cm².

2.2 Positron Trapping Model of a Single Type of Defect

The positron lifetime is defined as the time difference between the birth of a positron and the annihilation of positron. A thermalized positron in a perfect metal is annihilated with a constant annihilation rate $\lambda_b(=1/\tau_b, \tau_b=bulk$ lifetime). The positron lifetime of bulk Fe and Cr are known to be 106 ps and 118 ps, respectively [4]. In the presence of vacancy-type defects, positrons tend to trap at their sites with a trapping rate κ_d and annihilate with a annihilation rate λ_d (=1/ τ_d , τ_d =defect lifetime), which is schematically shown in Fig. 1. The positron decay function can be described as

$$f(t) = I_1 \exp(-\frac{t}{\tau_1}) + I_2 \exp(-\frac{t}{\tau_2})$$
(1)

where each lifetime and intensity are defined as

$$\begin{aligned} \tau_1 &= \frac{\tau_b}{1 + \kappa_d \tau_b}, \tau_2 = \tau_d, \\ I_1 &= 1 - I_2, I_2 = \frac{\kappa_d}{\lambda_f - \lambda_d + \kappa_d} \end{aligned} \tag{2}$$



Fig. 1. A schematic of the trapping model includes only one defect type.

From Eq. (2), the trapping rate κ_d can be calculated using experimentally measured PA data.

$$\kappa_{d} = (I_{2} / I_{1}) \cdot (\lambda_{b} - \lambda_{d})$$
(3)

According to the trapping model, the positron trapping probability is directly proportional to the defect concentration. Denoting a defect concentration C_d , the trapping rate κ_d is given as

$$\kappa_{\rm d} = \mu_{\rm d} \cdot C_{\rm d} \tag{4}$$

where the proportionality constant μ_d is the trapping coefficient.

3. Results and Discussion

The measured PA spectra are presented in Fig. 2. After an electron irradiation, the measured mean positron lifetime is slightly increased. We confirmed the creation of radiation induced point defects as a result of an electron irradiation. Based on these result, we assumed one defect type which we take here to be monovacancies.



Fig. 2. The measured positron lifetime spectra of the annealed Fe, and electron irradiated Fe-Cr alloys.

The positron lifetimes of a monovacancy in Fe and Cr are known to almost the same as 175 ps and 185 ps, respectively [5]. In the analysis of the PAS data, the intrinsic lifetime of an electron-irradiation induced defect (monovacancy) is set to be 175 ps, denoted by τ_d (= τ_2). The results for the positron annihilation lifetime data of the two samples are listed in Table I. As shown in Fig. 3, the single vacancy intensity (I₂) of the Fe-9Cr alloy is smaller than the other Fe-Cr alloys. This behavior is in good agreement with the previous irradiation resistance data [1,2].



Fig. 3. Measured lifetime and intensity. After an irradiation, the measured spectra were divided to two components.

Table I: Measured lifetime and intensity

Alloy	before	after electron irradiation			
	$\tau_b(ps)$	$\tau_1(ps)$	$I_1(\%)$	$\tau_2(ps)$	$I_2(\%)$
Fe	$106.1{\pm}0.2$	$100.5{\pm}0.6$	$91.43 {\pm} 0.49$	175(fix)	8.57±0.49
5Cr	111.2 ± 0.2	95.0±0.6	77.14±0.44	175(fix)	$22.86{\pm}0.44$
9Cr	111.2 ± 0.2	$102.0{\pm}0.6$	$83.86 {\pm} 0.53$	175(fix)	16.14±0.53
15Cr	111.2 ± 0.2	98.1±0.6	$78.49{\pm}0.48$	175(fix)	$21.51 {\pm} 0.48$

Then, we estimated the single vacancy concentration through Eqs. (3) & (4). We used the bulk positron lifetime (τ_b) of the Fe-Cr alloys from the measured positron lifetime of the well annealed Fe-Cr alloys. Then, the annihilation rate of the bulk ($\lambda_b=1/\tau_b$) and monovacancy ($\lambda_d=1/\tau_d$) were calculated. From these data, the positron trapping coefficients for vacancy were determined for the Fe-Cr alloys. The trapping coefficient for vacancies in pure Fe is known to be (1.1 \pm 0.2) x 10¹⁵/s from electron irradiation experiment [4]. Through the combination of the trapping rate and the trapping coefficient, the concentrations of monovacancy for the Fe-Cr alloys were estimated. The results are listed in Table II.

Table II: Problem Description

Alloy	trapping rate (κ_d , 10 ¹² /s)	Concentration (C _d , ppm)
Fe	3.480 ± 0.200	0.3164 ± 0.0603
5Cr	9.714 ± 0.200	0.8831 ± 0.1616
9Cr	6.247 ± 0.210	0.5679 ± 0.1050
15Cr	10.820 ± 0.228	0.9836 ± 0.1800

4. Conclusions

Through the application of positron annihilation measurement techniques, we investigated the radiationinduced point defects in Fe-Cr alloys. From the PALS, the formation of monovacancies in the irradiated materials was confirmed. Then we employed a positron trapping model of a single type of defect. By combining the trapping rate and trapping coefficient, the concentrations of the electron irradiation induced defects were estimated. From these results, we confirmed that Fe-9Cr alloy had better radiation resistance properties among Fe-Cr alloys. The application PAS provided information on radiationinduced defect production.

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