Investigation of Defect Concentration for ARAA using the Positron Annihilation Lifetime Spectroscopy

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1. Introduction

Advanced Reduced-Activation Alloy (ARAA) has been developed since 2011, as a structural material of fusion reactor [1]. A reduced activation ferritic-martensitic (RAFM) steel is being developed in many countries as a structural material for international thermonuclear experimental reactor (ITER) not only in Korea but also Europe (Eurofer), USA (9Cr-2WVTa), China (CLAM) and Japan (F82H), etc. ARAA, which will be applied to the test-blanket module of ITER, is placed in an environment where 14 MeV high energy neutron irradiation and tritium breeding occur. In case of nuclear structural materials can produced the point defect and cluster, which can lead to material degradation and cause problems with integrity of the facility. Therefore, ARAA must be able to endure in these extreme environment and it will be very important to know the information about the defects occurring in the structural materials.

There are several methods to identify defects or dislocation on thin surfaces, such as scanning electron microscope (SEM), transmission electron microscope (TEM), X-ray scatting, and scanning tunneling microscope (STM), atomic force microscope (AFM), and positron annihilation spectroscopy (PAS). Among them, PAS is known as a method for identifying microstructures at the thinnest depths using positrons as a probe [2]. The three main measurement techniques are associated with PAS. One is positron annihilation lifetime spectrum (PALS). Which can identify information of defect in material as measuring the lifetime of the positron. Another is Doppler Broadening Spectrum (DBS), which can determine the chemical characteristics of a substance by measuring the distribution of the electron momentum. The rest is Angular Correlation of Annihilation Radiation spectrum (ACAR), which can identify the electron structure of material by measuring the small angular deviation correlation in the decaying gamma quanta that occur with momentum of the electron. The radioisotope (RI) used PAS system gives us many information of material such as the presence of defects, the structural changes, and the dislocations of molecules, etc.

In this study, PALS was performed to identify defects according to the temperature and the rolling method of ARAA.

2. Experimental and Material

2.1 PALS

In this study, the Positron Annihilation Spectroscopy (PAS) was used for defect analysis of ARAA. PAS system consists of high voltage power supply, BaF_2 scintillators, photo-multiplier tubes (PMT), constant fraction differential discriminators (CFDD), nanosecond delay, multi-channel analyzer (MCA), and time to amplitude converter (TAC).

For optimization, we determined the suitable operating voltage for PMT and adjusted the hardware parts such as the two CFDD windows, the delay time, and the walk voltage.

The PAS uses about 50 μCi (about 1.85 MBq) foil-supported Na-22 radiation source, and the supporting material is 2.5 μm nickel foil. The source is placed between the samples, and the size is 8×8 mm^2. PALS more than $3x10^6$ counts were accumulated for each sample for 20,000 second. The time resolution is about 270 ps of full width at a half maximum (FWHM). The obtained spectra analysis was performed with the two Gaussian module which are resolutionfit and positronfit, in PALS*fit*3 package [3]. The fitted spectra all had a chi-square/dof value of less than 2.

2.2 Samples

We examined five kinds of ARAA. ARAA was created by adding 0.01 wt% Zr to 9Cr-1.2W based ferritic-martensitic steels. The composition of ARAA is given in Table 1. All samples were normalizing and rolling for the different method. The sample size was 10x10x1 mm^3. The specific sampling method for ARAA is described in Table 2.

Table I: The chemical composition of ARAA

Element	Content (wt %)	Element	Content (wt %)
Carbon	0.1	Vanadium	0.2
Silicon	0.1	Tantalum	0.07
Manganese	0.45	Nitrogen	0.01
Chromium	9	Titanium	0.01
Tungsten	1.2	Zirconium	0.01

Table II: Sampling method of ARAA

Sample	Annealing and Rolling method
VB27	Normalizing + Tempering (750 ℃/70 minute/air-cooling)
TMP2B	Normalizing + 25% cold-rolling
TMP13C	Normalizing
TMP34	Normalizing + 25% hot-rolling at 700 ℃
TMP38	Normalizing + 25% hot-rolling at 500 ℃

*Normalizing: 1000 ℃/40 minute/air-cooling
*TMP: Thermo-Mechanical Processing

3. Result

The longer lifetime (τ_2) which reflects the characteristics of the defect and mean lifetime was the longest for TMP34 sample, but intensity was the lowest. On the other hand, the longer lifetime was the shortest for TMP38 sample, but intensity was the highest. Because of this result, accordingly, the trapping rate (κ_d) was also the largest. The results for the positron annihilation lifetime of the five samples are shown in Table III.

Table III: The measured lifetime and intensity

ARAA	Lifetime components			
	τ_1 (ps)	I_1 (%)	τ_2 (ps)	I_2 (%)
VB27	141.5	83.8	498.8	16.2
TMP2B	146.9	80.1	469.6	19.9
TMP13C	139	81.1	503.1	18.9
TMP34	146.3	84.7	554.4	15.3
TMP38	135.9	79.5	449	20.5

The ARAA sample used in this experiment was estimated to generate defects during heat treatment and rolling process. In this study, the one defect type trapping model was applied to calculate the defect concentration [4]. In the applied model, the time dependent positron decay function is expressed as,

$$\mathrm{N(t)} = I_1 \exp(-\lambda_1 t) + I_2(-\lambda_2 t) \quad (1)$$

If the above equation is solved for the initial condition t=0 and $n_d(0)=0$, the following equation can be obtained.

$$\tau_1 = \frac{1}{\lambda_b + \kappa_d}, \quad \tau_2 = \frac{1}{\lambda_d}$$

$$I_1 = 1 - I_2, \quad I_2 = \frac{\kappa_d}{\lambda_b - \lambda_d + \kappa_d} \quad (2)$$

The value of the shorter lifetime (τ_1), the intensity of shorter lifetime (I_1) and the intensity of longer lifetime (I_2) can be used to calculate the trapping rate (κ_d) as the experimental result being proportional to the defect concentration.

$$\kappa_d = \mu_d C = I_2\left(\frac{1}{\tau_1} - \frac{1}{\tau_2}\right) = \frac{I_2}{I_1}\left(\frac{1}{\tau_b} - \frac{1}{\tau_d}\right) \quad (3)$$

Where μ_d is the trapping coefficient, and this value can vary by the type and state of the material. The most known value is (1.1×10^{15} /s) at temperature 200 K for vacancy of pure Fe, which was used to estimate the defect concentration in this study [5]. The result are shown in Table 4.

Table IV: The calculated defect concentration

ARAA	Defect concentration (C, ppm)
VB27	0.745
TMP2B	0.845
TMP13C	0.89
TMP34	0.69
TMP38	0.954

3. Conclusions

The defect ratio of all samples were analyzed using one defect type trapping model considering each coefficient. From the PALS, we confirmed the formation of defect in the samples. The analyzed defect ratio for all samples are affected by sample preparation process. It was confirmed that the defect of ARAA can be restored by heat treatment at approximately 700 ℃. It is expected that more accurate defect concentration can be calculated if the trapping rate and the exact trapping coefficient for the material are determined.

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