Investigation of Advanced Reduced-Activation Alloy (ARAA) with Coincidence Doppler Broadening Spectroscopy

Junyoung Lee [a,c], Jaegi Lee [a*], YoungSu Jeong [a],Gwang Min Sun [a], Young Rang Uhm [a], Young-Bum Chun [b], Sung-Joon Ye [c]
[a]*Neutron and Radioisotope Application Research Division, KAERI, 111 Daedeok-daero 989beon-gil, Daejeon, 34057*
[b]*Advanced Materials Research Division, KAERI, 111 Daedeok-daero 989beon-gil, Daejeon, 34057*
[c]*Program in Biomedical Radiation Sciences, Department of Transdisciplinary Studies, Graduate School of Convergence Science and Technology, Seoul National University, Seoul, 08826*
[*]*Corresponding author: jgl@kaeri.re.kr*

1. Introduction

Positrons injected into a dense matter almost instantly thermalized by the repulsive force from the nucleus. The energies of the positron annihilation photons are shifted by the Doppler effect, where the quantity of the gamma-ray energy shift is correlated to the momentum distribution of electrons. Nevertheless, the detection of annihilation gamma-rays from the core electrons is a challenging task, due to both the rarity of core electron annihilation events and the comparatively dominant Compton background signal acting as a noise. To enhance the signal-to-noise ratio of the energy spectrum of the annihilation gamma-rays, the coincidence Doppler broadening spectroscopy (CDBS) with two germanium detectors was introduced [1].

The reduced-activation ferritic-martensitic (RAFM) steels are promising candidates for structural material for the blanket of a nuclear fusion reactor. For the test blanket module of ITER, advanced reduced-activation alloy (ARAA) was developed in Korea as an alternative version of RAFM steels [2]. To enhance the properties of ARAA steels, thermo-mechanical processing (TMP) was applied to control the microstructure without modification of chemical composition [3]. Ausforming, one of the classes of TMP, is known to ameliorate the yield and tensile strengths of ferritic-martensitic steels. In this work, five ARAA samples were produced, each in a different TMP, and then were analyzed by the KAERI positron annihilation spectroscopy system.

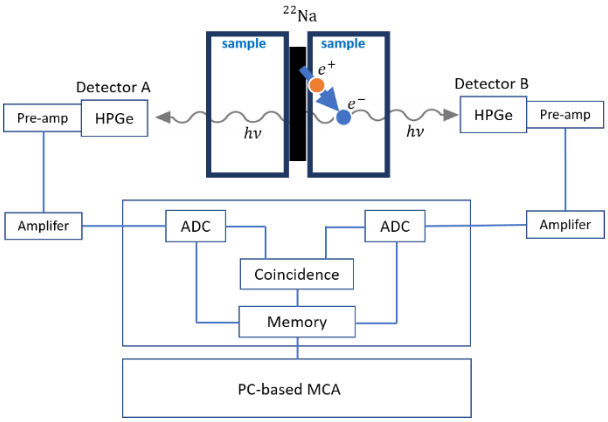
Table I: Chemical composition of ARAA (wt%)

C	Si	Mn	Cr	W
0.1	0.11	0.26	9.04	1.25
V	Ta	N	Ti	Zr
0.2	0.06	0.004	0.006	0.01

2. Methods and Results

2.1 System Setup

The KAERI coincidence Doppler broadening spectroscopy system consists of two HPGe detectors (Ortec GMX40P4-76), a dual 5-kV detector bias supply (Ortec 660), two amplifiers (Ortec 570), and a coincidence module (Labo NT24-DUAL). The coincidence module contains two analog-to-digital converters (ADC) with a maximum of 4,096 channels available and a coincidence timing module. The coincidence timing window was set to 1 μs. A 50 μCi ^{22}NaCl source was placed between 2.5-μm nickel foils. The distance between the HPGe detectors was set to 26 cm with a source in the middle, as the detectors were diametrically positioned. The shaping time of the amplifiers was 3 μs. ^{22}NaCl source was sandwiched between the samples. The direction angle of the samples to the HPGe detectors was 45° for simultaneous measurements of positron annihilation lifetime.



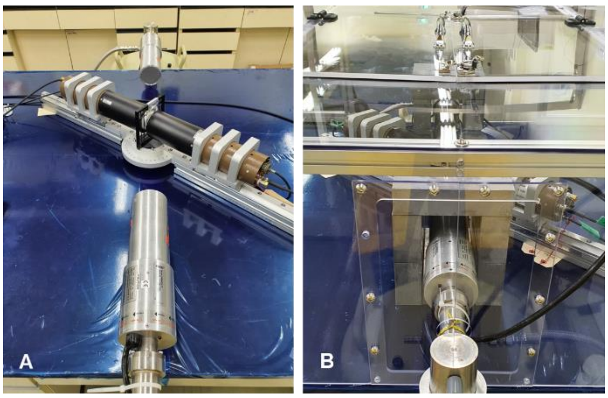


Fig. 1. Typical schematic diagram of coincidence Doppler broadening spectroscopy (CDBS) system. A) Radioisotope source and HPGe detector. B) Appearance of the CDBS system during measurement.

The ADC channels of two detectors were calibrated by ^{22}Na and ^{226}Ra sources. The four typical prompt gamma-rays from the ^{226}Ra source with energies of 186.1, 242.0, 295.2, and 351.9 keV as well as the 511 keV annihilation gamma-ray from ^{22}Na were counted to determine the channel-energy calibration curve.

Lead brick shields were placed around the active volume of the HPGe detector to reduce the background noise. An acrylic cage and a temperature controlling system were placed for the stability of measurement (Fig. 1.).

2.2 Test Specimen Preparation

To study the effect of TMP on mechanical properties, the specimens were austenitized and then subjected to five different TMPs as summarized in Table II. A reference plate, TMP13(C), was prepared by double normalizing and air-cooling for comparison.

Table II: TMP conditions of ARAA samples

TMP	TMP CONDITIONS
TMP13(C) (REF)	N*+N (double normalizing)
TMP21	N+N + 25% HR** at 600K
TMP38	N+N + 25% HR at 800K
TMP32	N+N + 15% HR at 1000K
TMP34	N+N + 25% HR at 1000K
TMP26	N+N + 25% HR at 1000K (25% hot-rolling in single pass)

N*: normalizing (1300K / 40 m / air-cooling)
HR**: hot-rolling

2.3 Experimental Result

The contour plot of the coincidence spectrum is shown in Fig. 2. The horizontal and the vertical bands correspond to the intensities of the annihilation gamma rays of the individual detector. The intense peak at the center matches with the counts for annihilation photons with an energy of 511 keV. The elliptical region extending diagonally, where the sum of two photons is equal to 1,022 keV, illustrates the pure Doppler shift.

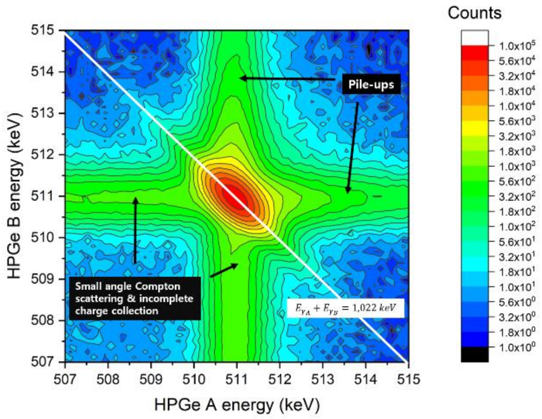


Fig. 2. Two-dimensional contour plot of the coincidence energy spectrum of the observed gamma-rays.

A self-developed Python algorithm was used to analyze DB and CDB spectra of five different ARAA specimens by applying the Gaussian and parabola combined model functions [4-5]. The CDB spectra, extracted by the algorithm, are shown in Fig. 3.

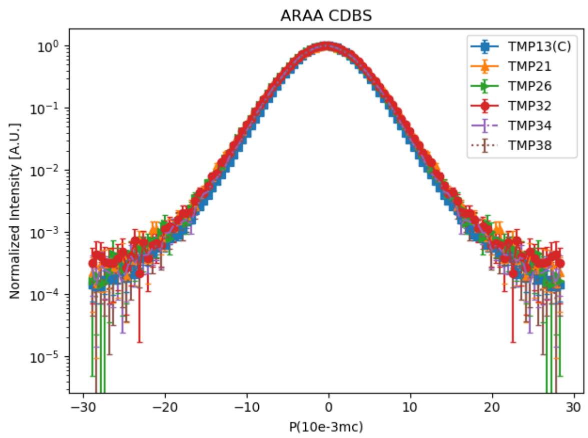


Fig. 3. Doppler broadening spectrum of ARAA sample measured with a single HPGe detector (blue dots) and coincidence Doppler broadening spectrum of the same sample obtained with two HPGe detectors in coincidence (orange triangle)

The Doppler shift (ΔE) can be translated as the electron momentum of the target as following Equation (1), where P_L is the electron momentum along the detector axis and c is the speed of light.

$$\Delta E = \frac{cP_L}{2} \quad (1)$$

Using a TMP13(C) as the reference sample, the CDBS ratio curves of ARAA samples was calculated. Fig. 4 illustrates the momentum ratio change due to the temperature of the hot-rolling process, whereas Fig. 5 shows the effect of various rolling conditions besides the temperature.

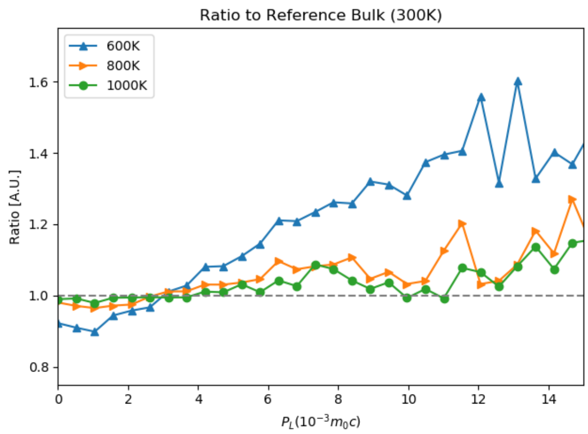


Fig. 4. The CDBS ratio curves of 25% HR ARAA samples with different hot-rolling temperature (TMP13(C) as a reference).

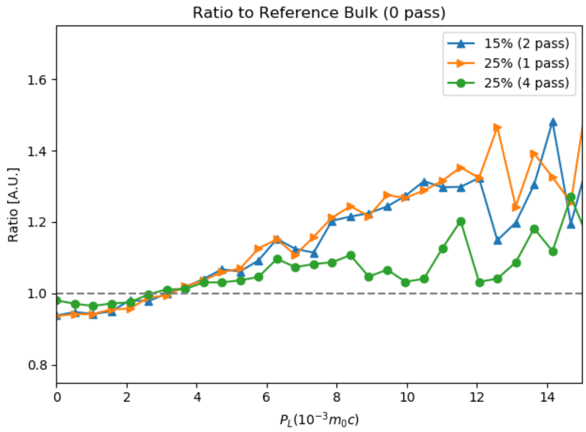


Fig. 5. The CDBS ratio curves of 1000K HR ARAA samples with different rolling conditions (TMP13(C) as a reference).

After the Gaussian curve fitting and normalization of the photon count spectrum, the S parameter was calculated for each Doppler shift (Table III).

Table III: S parameters according to TMP conditions

TMP CONDITIONS	S PARAMETER
N+N	0.5799
N+N + 25% HR AT 600K	0.5381
N+N + 25% HR AT 800K	0.5741
N+N + 15% HR AT 1000K	0.5501
N+N + 25% HR AT 1000K	0.5649
N+N + 25% HR AT 1000K (SINGLE PASS)	0.5524

S parameter and electron momentum do not directly indicate the mechanical properties but rather imply the fundamental structural properties such as lattice defects and electron momentum distribution within the matter.

3. Conclusions

Generally, a larger S parameter and a smaller ratio at the low electron momentum region can be interpreted as less lattice detect within the specimen compared to the reference sample. Both the electron momentum profile and the S parameter indicate that the lower the temperature of hot-rolling, the less microstructural defect the TMP sample possesses. In 1000K temperature fixed condition, the TMP of 15% hot rolling and 25% hot-rolling with single-pass showed negligible difference, whereas 25% hot-rolling with four passes have shown inferior scores.

Based on the positron annihilation spectroscopy results on different TMPs, hot rolling of 25% single pass at low temperature is recommended. However, further studies have to be conducted on the correlation of CDBS results and the mechanical properties of ARAA steels.

Acknowledgment

This work was supported by the National Research Foundation of Korea (NRF) Grant funded by the Korea government (MSIT) (NRF-2017M2A2A6A05018529).

REFERENCES

[1] K.G. Lynn, J.R. MacDonald, R.A. Boie, L.C. Feldman, J.D. Gabbe, M.F. Robbins, E. Bonderup, J. Golobchenko, Positron-Annhilation Momentum Porfiles in Aluminum: Core Contribution and the Independent-Particle Model, Physical Review Letters 38, p.241-244, 1977

[2] Y.B. Chun, S.H. Kang, D.W. Lee, S. Cho, Y.H. Jeong, A. Żywczak, C.K. Rhee, Development of Zr-containing advanced reduced-activation alloy (ARAA) as structural material for fusion reactors, Fusion Engineering and Design, Vol. 109-111, p.629-633, 2016

[3] Y.B. Chun, C.K. Rhee, D.W. Lee, Y.H. Park, Enhanced high-temperature mechanical properties of ARAA by thermos-mechanical processing, Fusion Engineering and Design, *Article in press*

[4] P. Asoka-Kumar, M. Alatalo. V.J. Ghosh, A.C. Kruseman, B. Nielsen, K.G. Lynn, Increased Elemental Specificity of Positron Annihilation Spectra, Physical Review Letters, Vol.77, p.2097-2100, 1996

[5] P. Pikart and C. Hugenschmidt, Novel data evaluation algorithm for Coincident Doppler Broadening Spectroscopy, Nuclear Instruments and Methods in Physics Research A, Vol.750, p.61-68, 2014.