[논 문] 한국재료학회지
Korean Journal of Materials Research
Vol. 3, No. 1 (1993)

최종열처리와 용접이 Zircaloy-4의 방사선조사 성장에 미치는 영향

임갑순 · 한정호 · 정용환 · 이덕현 · 박기성 · 김영석 · 김선진*
*한양대학교 재료공학과

The Effect of Final Heat Treatment and Welding on Irradiation Growth of Zircaloy-4

K.S. Rheem, J.H. Han, Y.H. Jeong, D.H. Lee, K.S. Park, Y.S. Kim and S.J. Kim*
Korea Atomic Energy Research Institute
**Dept. of Materials Eng., Hanyang Univ.*

초 록 최종열처리와 용접이 Zircaloy-4의 방사선조사 성장에 미치는 영향을 조사하였다. 본 연구에서는 중성자 조사에 대한 모의시험으로 3.5MeV로 가속된 양십자 빔을 조사량 $9.8 \times 10^{21} p/m^2$까지 시편에 조사하였다. 본연구에 사용된 시편중 annealed 시편의 방사선조사성장이 가장 컸으며 β-quenched 시편의 방사선조사 성장이 제일 작았다. 방사선조사 성장의 크기는 용접을 함에 따라 감소하였다. 최종열처리 조건의 차이에 의한 방사선조사 성장 크기에서의 차이와 용접이 방사선조사 성장에 미치는 영향을 X-ray 회절시험으로부터 계산된 Kearns number, f,를 이용하여 정량적으로 분석하였다.

Abstract The effect of final heat treatment and welding on the irradiation growth of Zircaloy-4 was investigated. As a simulation for neurtron irradiation, accelerated proton beam with the energy of 3.5MeV was used up to the proton fluence of $9.8 \times 10^{21} p/m^2$ in the present study. It was found that irradiation growth of the annealed specimen was the highest and that of the β-quenched specimen was the samllest among the present specimens. The magnitude of irradiation growth of the present specimens decreased by welding. The difference in the magnitude of irradiation growth of the present specimens with different final heat treatment and the effect of welding on it were quantitatively analyzed in terms of crystallographic texture by using Kearns number, f, which was calculated from the x-ray diffraction data.

I. Introduction

It is well known that zirconium and its alloy exhibit irradiation growth which is the dimensional change of material under irradiation in the absence of an applied stress[1]. According to the previous studies[2~4], irradiation growth is influenced by crystallographic texture, degree of cold work, neutron dose and temperature. A knowledge of the effects of these parameters on the irradiation growth behavior is important because the irradiation induced dimensional change of nuclear reactor components such as pressure tubes and the calandria tubes in the CANDU reactor and fuel cladding in all water cooled reactors can cause severe safety-related problems.

Among the parameters, the crystallogrphic texture is considered to have significant effect on irradiation growth[5]. Buckley[6] reported that zirconium single crystal expands along its a-axis and contracts along its c-axis without any overall volume change under irradiation. Therefore, it has been considered that irradiation growth of polycrystalline zirconium and its alloy is influenced significantly by the crystallographic texture in the growth direction[7~9]. Ibrahim and Winegar[10] found that the irradiation growth behavior of annealed zirconium alloys was in broad agreement with a texture model[11]. Recently Garzarolli et. al.[12] also reported the results of irradiation growth of zirconium alloys with different crystallographic texture due to different final treatments. They found that irradiation growth rate of β-quenched Zircalloy was 36% of that of fully recrystallized Zircaloy and it was in good agreement with the texture model. It is also well noted that crystallographic texture of zirconium alloy is changed by welding. During welding, weld zone of a zirconium alloy is transformed into β-quenched structure with randomized crystallographic texture. Accordingly, it is expected that irradiation growth behavior of weld zone of a zirconium will be different from that of the base zone due to the difference in crystallographic texture. The difference in the irradiation growth behavior between the weld zone and base zone can produce harmful residual stress which may cause safety-related problems in nuclear reactors. Therefore, it is needed to study the effect of welding on the irradiation growth behavior of zirconium alloys. Unfortunately, however, not much studies have been done on the effect of welding on the irradiation growth behavior of zirconium alloys. In the present study, the effect of welding on irradiation growth of Zircaloy-4 plates with different final treatments was investigated with the emphasis on crystallographic texture. As a simulation for the neutron irradiation, accelerated proton beam was employed in the present study. In light-ion irradiation such as proton irradiation, where the fraction of lower energy primary knock-on atom is large, the thermal spike effect is not so significant that the probability of defect survival is higher than in neutron irradiation.

Since only the surviving defects contribute to irradiation growth, more significant irradiation effects are expected in proton beam irradiation than in neutron irradiation and the simulation test facilities has the advantages in controlling of irradiation flux, tempreature, and stress compared to in-reactor experiments.

Ⅱ. Experimental Details

1. Specimen Preparation

In the present study, three kinds of specimens with different final heat treatments, i.e, as-received, annealed and β-quenched specimens were prepared from Zircaloy-4 plates with the thickness of 2mm which were purchased from Teledyne Wahchang Albany. The chemical composition of the Zircaloy-4 plate is shown in Table 1. The annealed specimens were prepared by heating the as-recieved Zircaloy-4 plates in a vacuum state of 10^{-3} torr for 24 hours at 900℃ before furnace colling in order for allowing grain growth and the β-quenched specimens were prepared by water quenching the as-received Zircaloy-4 plates after heating for 1 hr at 1000℃. After preparing the specimens, TIG welding was performed on each specimen by a bead-on-plate method in order to get weld structure on each specimen. From the welded specimens, the weld zone, heat affected zone and base zone were separated by cutting with a blade. Each separated specimen was mechanically ground and chemically thinned down to 0.1mm. From the thinned specimens, irradiation growth specimens of the size of 2mm×30mm×0.1mm were made by arc cutting and precision grinding. The microstructures of the specimens were observed under an optical microscope before proton irradiation.

Table 1. Chemical composition of the Zircaloy-4

Element	Sn	Fe	Cr	Zr
Wt%	1.53	0.20	0.11	Balance

*Impurities in ppm(O : 1380, C : 170, H : 6)

2. Crystallographic Texture Analysis

In general crystallographic texture has been characterized by using a direct pole figure or an inverse pole figure from X-ray diffraction data. However, it has been difficult to compare direct pole figures or inverse pole figures quantitatively. In the meantime, various texture numbers were defined in order to allow a quantitative comparison[13~15]. Of the texture numbers, f parameter defined by Kearns[16], which is widely used in the nuclear industry, was used in the present study to characterize the crystallographic texture of the specimens. The f parameter is given by

$$f = \int I_{\phi} \sin\phi \cos^2\phi \, d\phi \qquad (1)$$

where f = effective fraction of cells aligned with their certain axis parallel to the reference direction

I_{ϕ} = average pole density at an angle ϕ from the reference direction

ϕ = tilt angle from the reference direction.

The f values for the basal pole (0002) at the plane perpendicular to the rolling direction, which is irradiation growth direction of the specimens were obtained from the X-ray diffraction data of the weld zone and base zone of each specimen. The details of the calculation for f values are given elsewhere[16]. X-ray diffraction experiment was done on the plane perpendicular to the rolling direction of specimens and Cu was used as a target material. The diffraction angle, 2θ, ranges was from 30° to 130° and the scan speed was 0.085°/sec.

3. Proton Irradiation

In the present study, neutron irradiation was simulated by using 3.5MeV proton beam accelerated in a 5SDH-2 Pelletron accelerator made by National Electostatics Corporation. A schematic illustration of the proton irradiaion set-up is shown in Fig. 1. The accelerated proton beam was collimated by an Al collimator which has a rectangular shape hole of the size of 4mm × 20mm. By using the collimated proton beam, the specimen, held in the specimen holder, was irradiated in a vacuum state of 10^{-6}torr. The current of the proton beam was measured through a Faraday cup and it was 3.5μA. The proton irradiation time was up to 450 minutes and the corresponding proton fluence was 9.8×10^{21}p/m^2. The temperature of the specimen during irradiation was measured with a calibrated Chromel-Alumel thermocouple and it was 370℃. The change in length of the specimens due to proton irradiation was checked by measuring the length of the specimens before and after proton irradiation with a He-Ne laser interferometer with the precision of ±0.1μm.

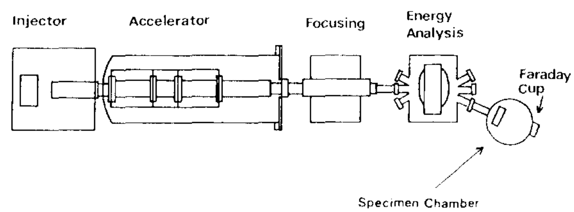

Fig. 1. Schematic arrangement for proton irradiation test.

4. Calculation of dpa

The value of displacement per atom(dpa), which represents the number of displacements of an atom from its original lattics site by irradiation of a beam having certain energy, is given by

$$dpa = \sigma_d \times fluence \qquad (2)$$

where σ_d is an energy-dependent displacement cross section of material. This value for neutron and electron is reported[17,18], but it could not get in the case of proton. Therefore, in present study the value of dpa was calculated with a beam flux and displacement numbers obtained from TRIM-90 computer code.

Ⅲ. Results and Discussion

1. Microstructure and Crystallographic Texture

Microstructures of base zones and weld zones of the present specimens are shown in Fig. 2 and the corresponding texture parameter f values, Kearns number, are presented in

Table 2. As shown in Fig. 2, the grains of the as-received specimen and the annealed specimen were equiaxed with the average grain sizes of 10μm and 20μm respectievly. In the mean time, the β-quenched specimen showed a well developed Widmanstätten structure which is composed of α plates in the prior β grains. The f values for the (0002) basal pole corresponding to the base zones of the as-received, annealed and β-quenched specimens were 0.270, 0.198 and 0.378 respectively as shown in Table 2. It means that the fraction of grains whose (0002) basal pole is oriented along the rolling direction of the as-received and annealed specimens is smaller than that of the randomized specimens whose f value is 0.333. However, in case of the β-quenched specimen, the fraction was somewhat larger than that would be expected for the randomized specimen.

Upon welding by bead-on-plate method, the equiaxed grains of the as-received and anneled specimens changed into Widmanstätten structures and the fraction of grains whose (0002) basal pole is oriented along the rolling direction of the specimens increased as shown in Fig.2 and Table 2.

2. Irradiation Growth

The results of irradiation growth at the proton fluence of about 9.8×10^{21}p/m^2 are presented in Fig.3. The length changes due to irradiation growth of the as-received, annealed and β-quenched specimens were 0.7μm, 2.82μm and 0.38μm and the corresponding strains were 3.5×10^{-4}, 14.1×10^{-4} and 1.9×10^{-4} respectively. Since the precision of the He-Ne laser interferometer was $\pm0.1\mu$m, the measured value of length change so the values of strain are considered significant. According to Holt[19], the magnitude of irradiation growth increases as grains are elongated along the irradiation growth direction. However, as one can see in Fig2, the grains of the as-received and annealed specimens are equiaxed. Therefore, effect of grain shape on the magnitude of irradiation growth is not expected for both of the specimens. Also for the β-quenched specimen, effect of grain shape is not expected even though the grain shape is not equiaxed because the α plates are distributed randomly. Therefore, the difference in the magnitude of irradiation growth of the present specimens must be due to the difference in crystallogrphic texture. According to Hesketh et. al.[20], the magnitude of irradiation growth of Zirconium alloys depends on crystallographic texture of the specimen and the dependence is commonly expressed as follows under the assumption of zero volume change during irradiation growth,

$$G_d = 1 - 3f_d \quad (3)$$

where G_d is a growth coefficient and f_d is a texture parameter for the basal pole (0002) in the growth direction d.

As expected from the f values for the present specimens and the eq.(3), the magnitude of irradiation growth of the annealed specimen was largest and that of the β-quenched specimen was the smallest. However, the present results showed some deviation from the (1-3f) dependence. According to the eq.(3), the magnitude, based on the as-received specimen, of irradiation growth of the annealed and β-quenched specimens should be about 7.5×10^{-4} and -2.5×10^{-4}(contraction), respectively. Rogerson and Murgatroyd[21] reported that the (1-3f) relationship does not apply at low dose of neutron($<1\times10^{24}$n/m^2) in cold worked Zircoloy-2 with high texture parameter. They thought that the deviation from the (1-3f) dependence must be due to volume change, which produce a complex crystallographic texture dependence of irradiation growth, at low neutron dose. In Fig. 4, the result of the TRIM code calculation for the displacement of Zr at the proton beam energy of 3.5 MeV is presented. The maximum displacement was about 35×10^3/ion · cm and the average value was about 5×10^3/ion · cm. By using the values and data of the present test conditions, i.e, pro-

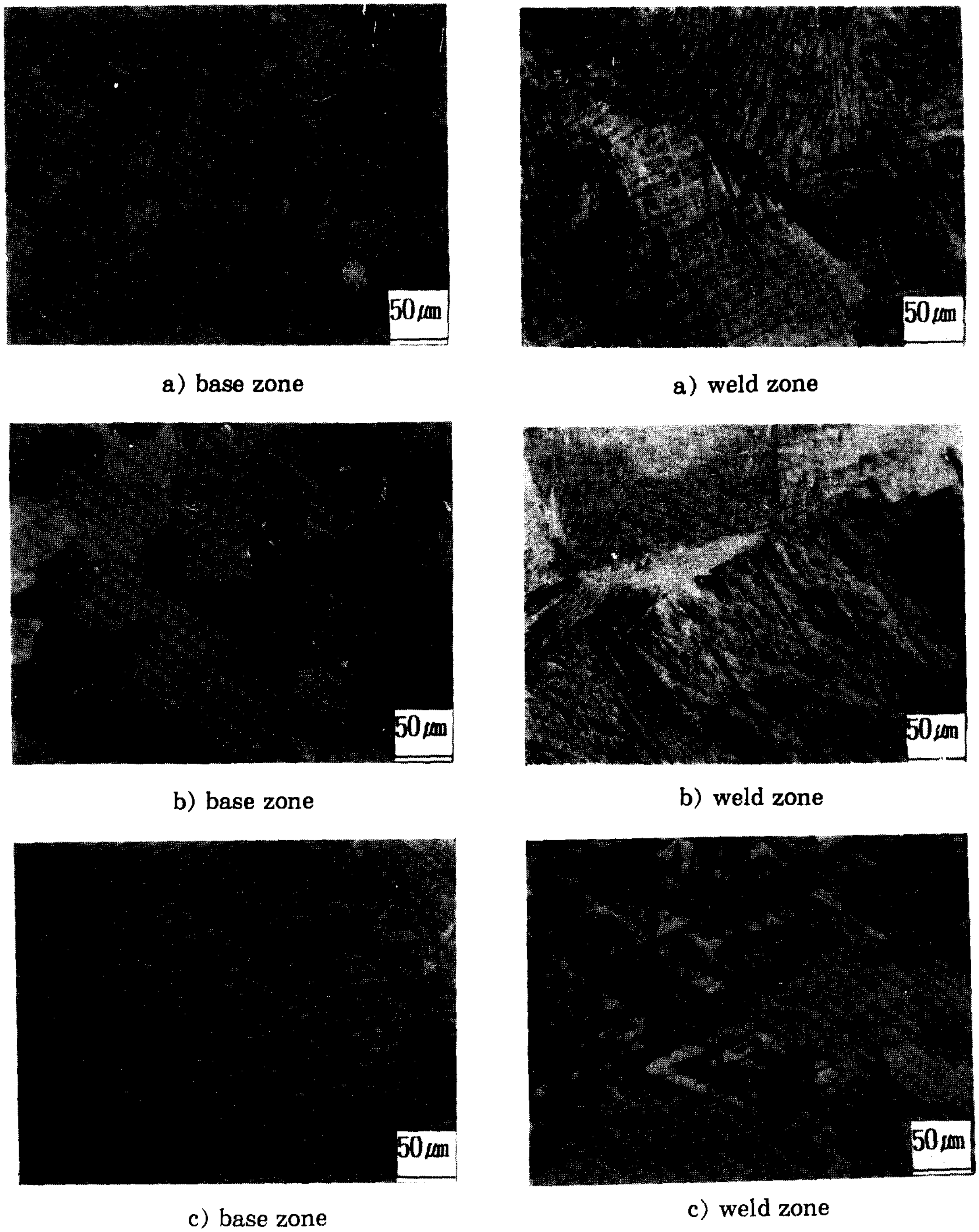

Fig. 2. Microstructures of the a) as received, b) annealed, c) β-quenched Zircaloy-4 plates

ton flux of 3.64×10^{3}p/ion · cm and irradiation time of 450 minutes, the dpa values were calculated. The maximum dpa was 0.8 and the average dpa was about 0.12. The corresponding neutron fluences were 7×10^{24}n/m^2 and 9 $\times 10^{23}$n/m^2 respectively. Therefore, it is considered that the deviation of the present results from the(1-3f) dependence could be partly due to the low proton fluence. Even though the present results showed some deviation from

Table 2. f parameter values of Zircaloy-4 plates with different final treatments

Specimen	f Parameter Value	
	Base Zone	Welded Zone
As-received	0.270	0.308
Annealed	0.198	0.377
β-Quenched	0.378	0.390

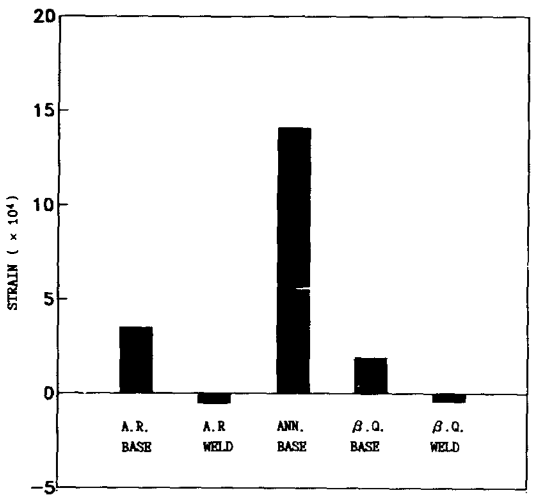

Fig.3. Irradiation growth of Zircaloy-4 with different final treatments and welding at the proton flunce of 9.8×10^{21}p/m^2.

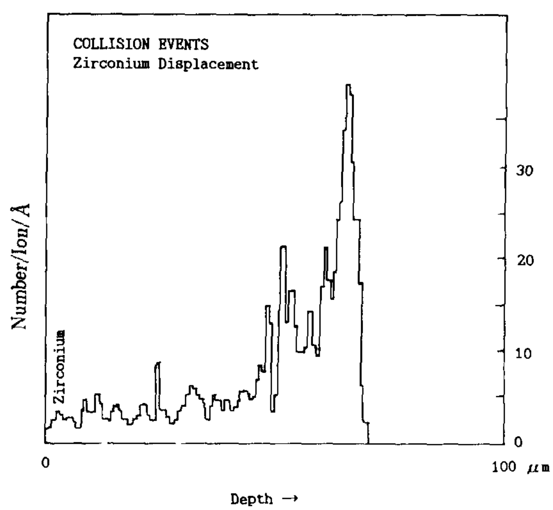

Fig.4. Displacement of Zirconium calculated by TRIM code for proton beam irradiation with the energy of 3.5MeV.

the (1-3f) dependence, the trend that the magnitude of irradiation growth of the β-quenched specimen is the smallest among the present specimens was in good agreement with the results obatined at higher·neutron fluence[22].

3. Effect of Welding

The length changes due to irradiation growth were decreased by welding and those of the weld zones of the as-received and β-quenched specimens were -0.1μm and -0.08μm and the corresponding values of strain were -0.5×10^{-4} and -0.4×10^{-4} respectively. Actually these measured values were out of the precision range of the He-Ne laser interferometer so that they are not reliable but it can be considered that the length changes so the values of strain must be very small. One thing to note is that the weld zones of the specimens contracted due to proton irradiation. It is considered to be due to the increase of f values by welding. As shown in Table 2, f values were increased by welding so that the f values of the weld zones of the as-received, annealed and β-quenched specimens were 0.308, 0.377 and 0.390 respectively. With these f values and the eq.(3), the contraction of the specimesn could be expected. Since the magnitude of irradiation growth of the weld zones of the specimens is considered negligibly samll, the difference in the magnitude of irradiation growth between the weld zone and base zone is the smallest in the β-quenched specimen. Thus, the samllest residual stress is expected to be built up in the β-quenched specimen during welding and the same is expected to be true at higher proton fluence.

CONCLUSIONS

1. The magnitude of irradiation growth was the largest in the annealed specimen and the smallest in the β-quenched specimen. The difference was considered to be due to the difference in the crystallographic texture.
2. The crystallographic texture parameter, f,

values for the as-received, annealed and the β-quenched specimens were 0.270, 0.198 and 0.378 respectively and they were increased to 0.308, 0.377 and 0.390 respectively by welding.

3. As expcted from the relation between the f value and the growth coefficient, the weld zones of the present specimens were contracted by proton beam irradiation. However, the magnitude was negligibly small. The growth strains of the weld zones of the as-received and β-quenched specimens were -0.5×10^{-4} and -0.4×10^{-4} respectively at the proton fluence of $9.8 \times 10^{21} p/m^2$.
4. The difference in growth strain between the weld zone and base zone was the smallest in the β-quenched specimen.

References

1. V. Fidleris, Atomic Energy Rev., 13, 51 (1975).
2. R.G. Fleck, Can. Metal. Q., 18, 65 (1979)
3. E.F. Ibrahim et al., J. Nucl. Mater., 91, 31 (1989).
4. R. G. Fleck et al., ASTM STP 824, 88 (1982)
5. R.B. Adamson et al., "High Temperature Irradiation Growth in Zircaloy", ASTM Zirconium Conference, Boston, August 4, 240(1980).
6. S.N. Buckley, "Properties of Reactor Materials and the Effects of Irradiation Damage", Butterworths, London, 413 (1961).
7. R.C. Daniel, Nucl Tech., 14, 171 (1972).
8. R.B. Adamson, ASTM STP 633, 326 (1977).
9. R.V. Hesketh et al., "Radiation Damage in Reactor Materials 1", IAEA, Vienna, 365 (1969)
10. E.F. Ibrahim and J.E. Winegar, J. Nucl. Mater., 45, 335 (1972).
11. J.E. Harbottle, ASTM STP 484, 287 (1970).
12. F. Garzarolli et al., "Properties of Materials for Water Reactor Fuel Elements and Methods of Measurements", IAEA, Vienna, 44 (1986).
13. K. Kallstrom, Can. Met. Quart., 11, 185 (1972).
14. K. Kallstrom, T. Anderson and a. Hofvenstram, ASTM STP 551, 160 (1974).
15. B.D. Knorr and R.M. Pelloux, J. Nucl. Mater., 77, 1 (1977).
16. J.J. Kearns, Westinghouse Co. Report WAPD-TM-472(1965).
17. D.G. Doran et al., ASTM STP 611 (1976) 463
18. O. S. Oen, ORNL-4897, Aug., (1973).
19. R.A. Holt, J. Nucl. Mater., 82, 419 (1968).
20. R.V. Hesketh, J.E. Harbottle, N.A. Waterman and R.C. Lobb., "Radiation Damage in Reactor Materials Vol 1". IAEA, Vienna, (1969).
21. A. Rogerson and R.A. Murgatroyd, J. Nucl. Mater., 80, 253 (1979).
22. Private Communication with Dr. Weidinger at KWU (1987).