Post-irradiation Examination Using TEM Method for Swelling Evaluation of Baffle Plate in PWR Core Internals

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Abstract

Irradiation Assisted Stress Corrosion Cracking (IASCC) occurs in core internal structures that receive high neutron irradiation in a pressurized water reactor (PWR) environment. In order to obtain the void swelling data of Type 304 stainless steel which is necessary for reliability evaluation of baffle structure in core internals, it is considered to be important to obtain data using baffle plate material sampled from the actual plant.

However, it is difficult to obtain the baffle plate directly from the actual plant under service in the effort of the post-irradiation examination (PIE). It was common to evaluate using materials sampled from baffle former bolts or flux thimble tubes. It is relatively close to the core, and the same kind of austenitic stainless steel (cold-worked Type 316 stainless steel) as baffle plate. Therefore, in order to improve measurement accuracy and reliability in the swelling evaluation of the baffle plate, it is the same material type, and it is desired to develop systematic data on the PWR irradiation conditions (dose, dose rate, and irradiation temperature) and swelling characteristic.

In this study, directly evaluation was made possible by conducting the PIE using the sampling of the irradiation material from the decommissioning plant. In order to obtain the systematic swelling data against irradiation conditions, it was taken from a baffle plate of the Spain’s decommissioning ZORITA PWR plant after 40Years (26EFPY) operation.

Swelling data of the baffle plate was obtained using transmission electron microscope (TEM) method with sampling technique from micro region by PIE in hot laboratory as follows:

- Based on the simulated analysis results of the dose and the irradiation temperature, the sampling position was selected using the current swelling evaluation formula. A size of 12mm×36mm×29mm (thickness) from the actual baffle plate was obtained.
- Swelling evaluated by TEM method, using dose (33~47dpa) and irradiation temperature (299~327°C) as parameters with PIE in Nuclear Development Cooperation (NDC) hot laboratory.
- In order to reduce radiation exposure to the human body due to radioactivity by TEM observation in the hot laboratory, processing was punched out in a micro region at φ1mm from irradiation material, embedded in an un-irradiated stainless steel material of φ3mm, and the thin film was processed by electrically polishing methods for high magnification cavity observation.
- As a result, a swelling ratio of 0.015%~0.080% was obtained based on irradiation conditions. It was possible to obtain the validity of the current swelling evaluation equation.

1. INTRODUCTION

Damage to baffle former bolts (BFBs) by Irradiation Assisted Stress Corrosion Cracking (IASCC) has become obvious in Pressurized Water Reactor (PWR) plants. It is an important issue for plant conservation with regard to BFB's IASCC in the world.

In the evaluation of IASCC for BFBs, in order to evaluate the susceptibility and initiation of IASCC accompanying neutron irradiation, and to evaluate the change with respect to the tightening stress on bolt, stress evaluation in consideration of the irradiation effect such as change in mechanical characteristics, void swelling, and relaxation must be done. It is known that swelling deformation (volumetric expansion) greatly affects the stress of BFBs for the Type 304 stainless steel baffle plate fastened by the BFBs shown in Fig.1 [1].

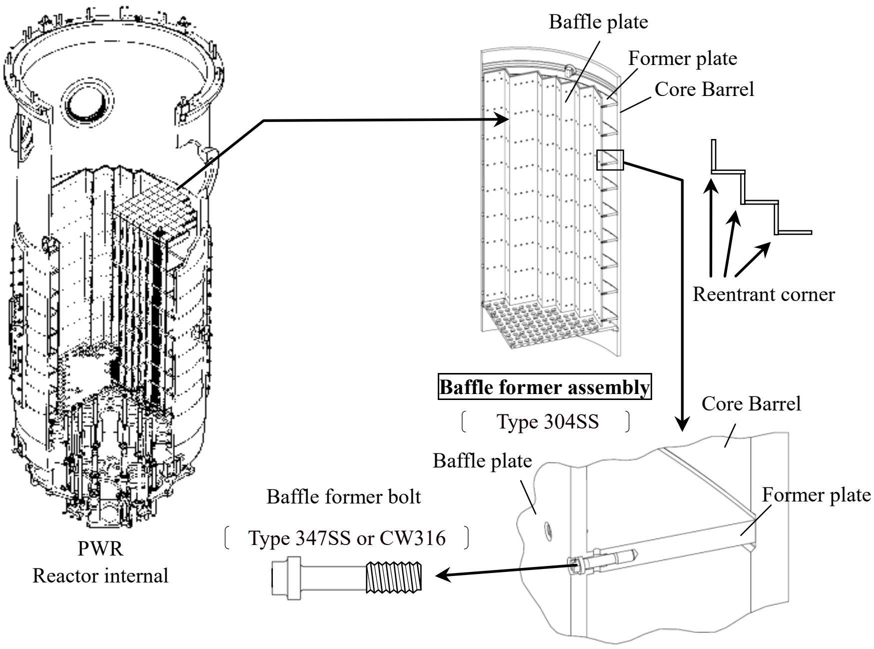


Fig.1 Configuration of baffle former assembly in PWR core internals

However, for swelling, the void swelling evaluation equation improved to PWR based on the data which has been modified in the Fast Breeder Reactor (FBR) EBR-II in the past (modified dpa rate corrected Foster-Flinn equation) [2, 3] has been under consideration. In order to evaluate the actual PWR plant, data acquisition using BFBs which is the steel type and cold-worked Type 316 stainless steel of flux thimble tube as the post irradiation examination (PIE) is under way. However, the current situation is that direct evaluation has not been made due to the kind of same steel and irradiation conditions (dose, dose rate, and irradiation temperature).

Therefore, in this research, as a part of the research using the Spanish decommissioned material conducted Zorita Internals Research Project (ZIRP) of the Electric Power Research Institute (EPRI), the results of analysis of dose and irradiation temperature based on the hot laboratory of Nuclear Development Corporation (NDC) were used, micro sampling from a baffle plate was carried out and data expansion by swelling measurement using a Transmission Electron Microscope (TEM) systematically was obtained. And the validity of the swelling evaluation equation was verified.

2. EXPERIMENTAL PROCEDURE

(1) Material

In this study, baffle plate was collected from the baffle structure of the reactor internal structure of the ZORITA decommissioning plant in Spain after about 40Years (26EFPY) operation, and based on analysis results of dose and irradiation temperature provided from EPRI described later. The sampling position was selected in EPRI ZIRP as a test coupon at the Studsvik laboratory in Sweden and then transported to NDC by Type A cask. The steel is Type 304 austenitic stainless steel, but there is no mill sheet data (chemical composition).

In the NDC, one obtained by processing to a size of 12mm×36mm×29mm^t (plate thickness) shown in the processing position from the baffle plate shown in Fig.2 and the appearance state of the test coupon (B1A2) in Fig.3.

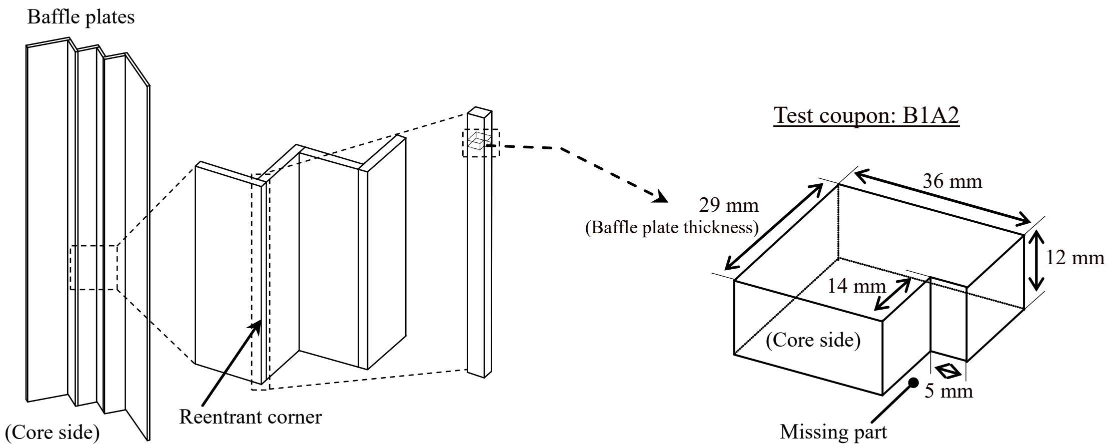


Fig.2 Schematic illustration of the test coupon removed from reentrant corner of baffle plate

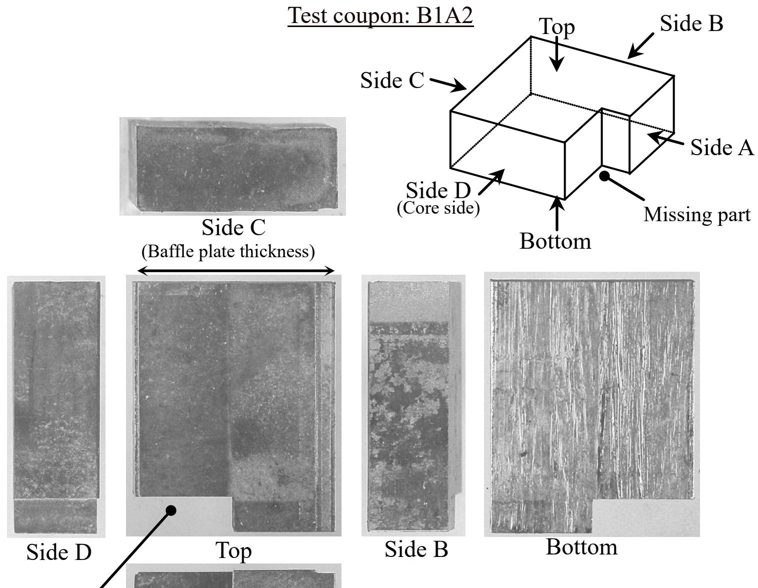


Fig.3 Pictures of the test coupon removed from reentrant corner of baffle plate

(2) Sampling Position of the Test Coupon

For the test coupon of 12mm×36mm×29mm^{t}, analysis results of irradiation dose and irradiation temperature provided from EPRI as shown in Fig.4 were referred to. In other words, in order to obtain the swelling ratio by the swelling evaluation equation, based on the analysis result, in order to expand the systematic swelling data and verify the validity of the swelling evaluation equation, the measurement position is systematically. Furthermore, in order to expand irradiation data of Type 304 stainless steel, three representative positions were decided.

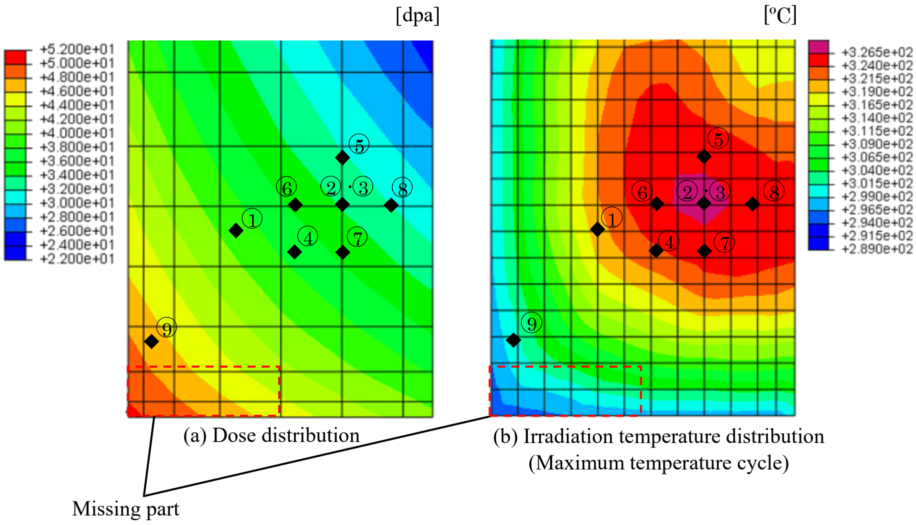


Fig.4 Calculated dose and irradiation temperature of the test coupon and sampling positions on the top side

As shown in Table 1, sample No. 1 is a position at which the maximum irradiation amount is obtained with respect to the irradiation temperature range of about 320°C or more. Sample No. 2 and 3 are the highest temperature. Sample No. 4 is a position intermediate between sample No. 1 and No. 2 or No. 3. Sample No.5~No.8 are the positions for expanding data around sample No. 2 or No. 3. Sample No. 9 was systematically selected as the position at which the maximum irradiation dose was obtained in the all-region.

Table 1 Sampling positions on the top side and irradiation condition from the test coupon

No.	Sampling positions on the top side	Dose (dpa)	Dose rate (dpa/s)	Irrad.Temp. (°C)
①	Max. dose position (over 320°C region)	39	4.8x10^{-8}	319
②	Max. temp. position	34	4.1x10^{-8}	327
③	Max. temp. position as same as No.② position	35	4.3x10^{-8}	327
④	Middle of No.① and No.② position	37	4.5x10^{-8}	324
⑤	Near max. temp. No.② position	33	4.0x10^{-8}	326
⑥	Near max. temp. No.② position	36	4.4x10^{-8}	325
⑦	Near max. temp. No.② position	36	4.4x10^{-8}	325
⑧	Near max. temp. No.② position	33	4.0x10^{-8}	326
⑨	Max. dose position (all region)	47	5.7x10^{-8}	299

(3) Samples Preparation

Fig.5 shows the processing position and procedure of the disk sample for TEM observation. 4mm×4mm×12mm^{L} was cut out from the test coupon with a hot laboratory of NDC. For disk samples for TEM observation, sliced into 4mm×4mm×1mm^t and finished to a thickness of about 70µm using emery abrasive paper up to #300~#1,200. In order to reduce radiation exposure due to radioactivity during TEM observation, punching to φ1mm size was performed. Furthermore, an irradiation material of φ1 mm in size was embedded in the center of the ring of φ3mm un-irradiated stainless steel, and after the Ni plating (Solution: $NiSO_4$+$NiCl_2$+H_3BO_3+pure water, Temperature: 60°C., Current density: 90mA/cm^2, Time: 20min), a hole was formed by twin-jet electrolytically polishing (Solution: 7% Acetic perchloric acid, Temperature: 11°C, Voltage: 30V) was tested.

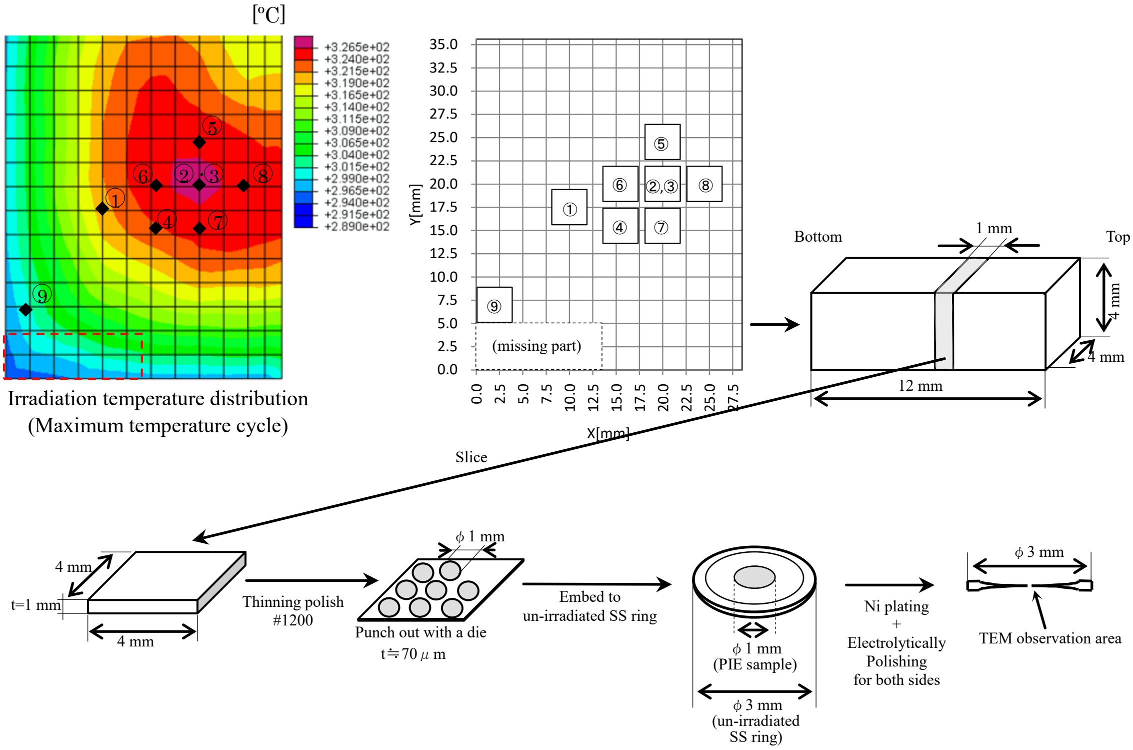


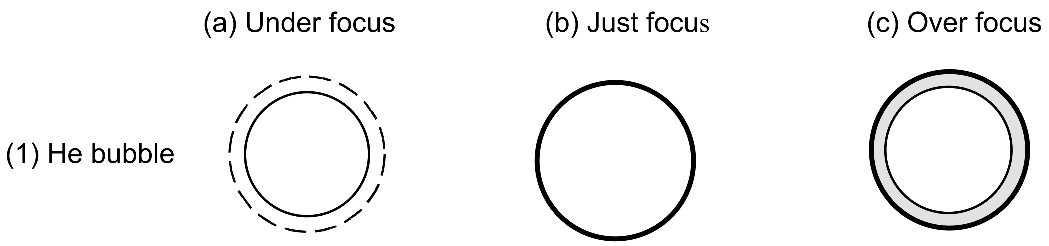
Fig.5 Preparation procedure of TEM disk samples

(4) Post-irradiation Examination

In order to perform swelling measurement, cavity observation was performed at high magnification in the grain using the TEM method. For the TEM observation, a JEOL JEM-2010F instrument with an acceleration voltage of 200kV was used. The number of repetition was changed from 9 samples to 3 parts, and a total of 27 sites were measured. The magnification in TEM observation was 100,000 times, 300,000 times, and 500,000 times.

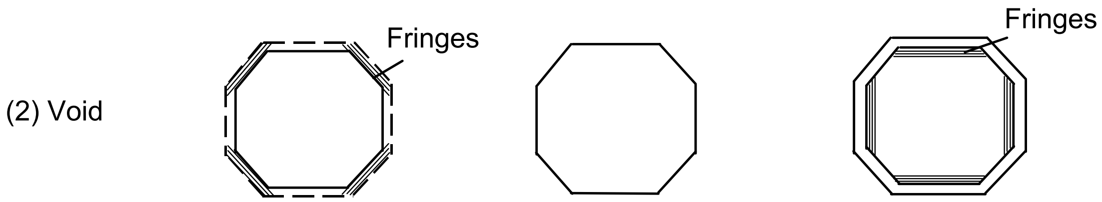
Generally, cavities are recognized as bubbles type filled with gas and voids type [4]. The bubbles have spherical shape, are filled with hydrogen or helium gas. And it is thought to be caused by transmutation of nickel element in the stainless steel. It is call helium bubbles. On the other hand, the voids have crystallographic polygonal shape having fringes, and it is thought that the void has a larger diameter than the helium bubble and greatly contributes to volumetric expansion of materials.

In observing these cavities, in order to distinguish between helium bubbles and voids, decided to change the focus point during TEM observation to over focus, just focus, and under focus are shown in Fig.6. That is, with respect to helium bubbles, the diameter size is relatively small, 1~3nm, and observation in a ring state accompanying a change in focus from a spherical shape. For voids, it is a diameter size of 3nm or more, tried to distinguish from polygonal observation.



In case of helium babble (cavity diameter: ≤3nm), the shape does not change even at each focus position, and it looks like a sphere shape

- ✓ At under focus, it looks like a white sphere shape
- ✓ At over focus, it looks like a black sphere shape



In case of void (cavity diameter: >3nm), different shape at each focus position, polygonal shape and fringes visible

- ✓ At under focus, it looks like a white polygonal shape and external bright fringes
- ✓ At over focus, it looks like a black polygonal shape and internal bright fringes

Fig.6 Image of the distinction between helium bubbles and voids

Swelling was measured by obtaining the diameter and the number of the cavity in the observation field of each sample. It is calculated by using the following swelling measuring equation (1) for obtaining the swelling ratio. The sample average thickness t at the TEM observation position was obtained by using Convergent-Beam Electron Diffraction (CBED) method [5].

$$S = \sum_{i=1}^{n} \left\{ \frac{4}{3}\pi \left(\frac{d_i}{2} \right)^3 \right\} \Big/ (At) \times 100 \quad \text{.....................} \quad (1)$$

Where,

S : swelling ratio [%]
n : number of cavities per observed area [-]
d_i : diameter of a cavity [nm]
A : observed area [nm^2]
t : average thickness of observed area [nm]

3. RESULTS AND DISCUSSIONS

(1) Swelling Measurement

Representative TEM pictures under focus for samples No.1~No.9 are shown in Fig.7. In the sample No.9 at the low irradiation temperature (299°C), the density was low, and the small diameter cavity size of about several nanometers was observed. The small diameter cavity was judged to be a helium bubble from the size and shape. On the other hand, in sample No.1 and No.2 at irradiation temperatures of high (319°C, 327°C, respectively), not only cavities of small diameter size of about several nanometer, but also cavities of large diameter of maximum about 8nm were formed. The sample No.9, this small diameter cavity size was judged to be a helium bubble. Regarding this large diameter cavity size, it was judged to be void from size and polygonal shape. As the irradiation temperature increases, believe that growth into voids was observed due to the trapping of thermally activated atomic vacancies.

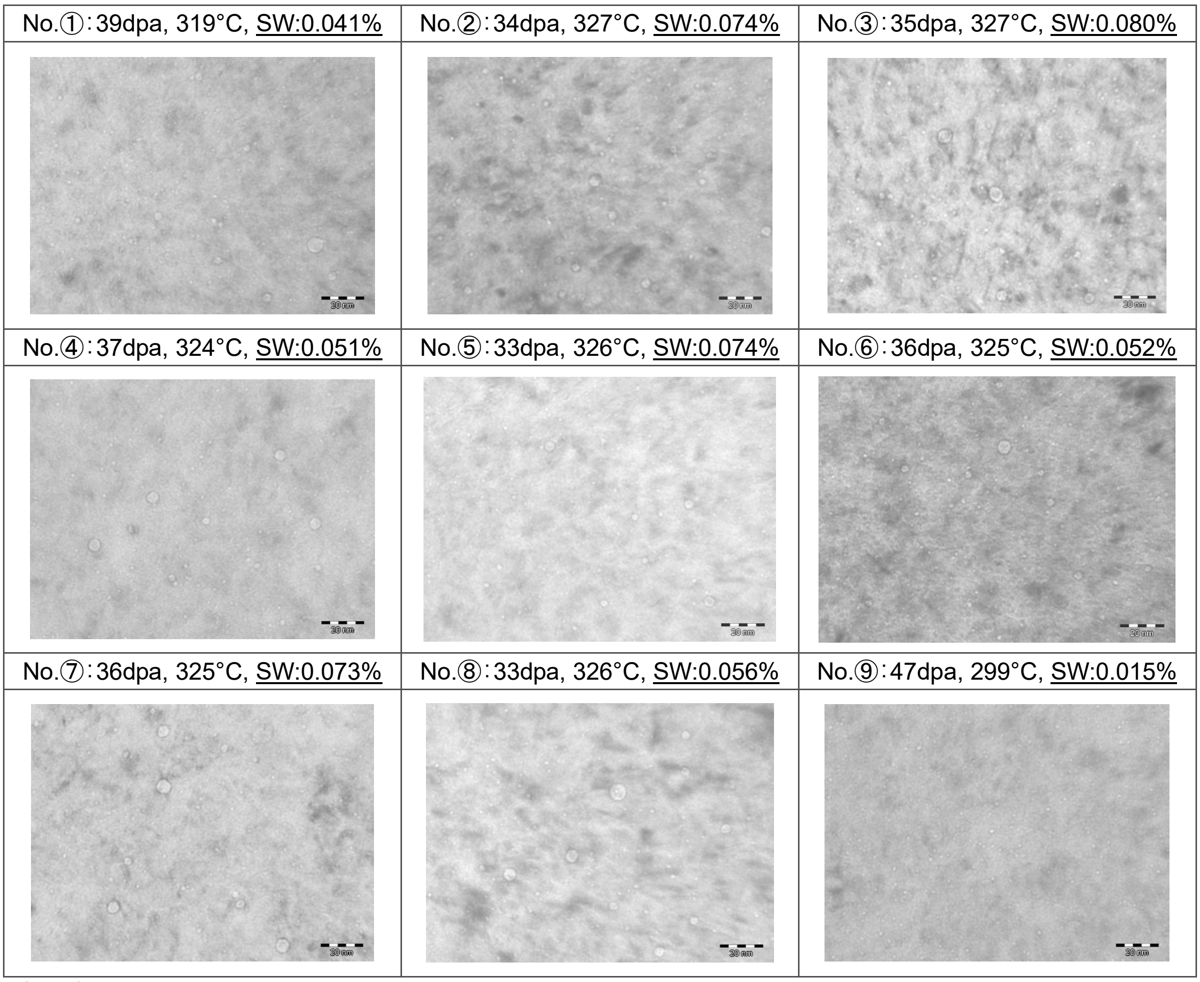
Results of the diameter size distribution for helium bubbles and voids are shown in Fig.8. For the distribution in the histogram, it is considered that the distribution is assumed to be a helium bubble of small diameter up to 3nm and the void is larger than 3nm in size, and it is classified and organized. Comparing sample No.1 with sample No.2, the distribution of the histogram of the helium bubbles tended to decrease as the irradiation temperature increased. On the other hand, the distribution of the histogram for voids larger than 3nm in size is thought to have grown from helium bubbles to voids at around 320°C as a boundary.

The observation results of the cavities obtained from these results and the results of calculating the swelling ratio based on the swelling measuring equation (1) are shown in Table 2. The maximum swelling ratio was 0.080% in sample No.3 with the highest irradiation temperature. The minimum swelling ratio was 0.015% in sample No.9, which had the lowest irradiation temperature.

In addition, Fig.9, Fig.10, and Fig.11 show the relationship between the irradiation temperature and dose, and dose rate for the swelling ratio obtained in this study, respectively. Also, Fig.12 shows the result of arranging the relationship between the swelling ratio and the irradiation temperature with respect to the literature data [6-12] of BFB and flux thimble tube of cold-worked Type 316 stainless steel irradiated in PWR in contrast to Type 304 stainless steel baffle plates in this study. Fig.13 similarly shows the results of arranging the relation with the dose.

As shown in Fig.9, the correlation with the swelling ratio in relation to the irradiation temperature was observed, and the tendency that the swelling ratio became larger as the temperature increased was observed. In a certain document [13], it has been reported that the swelling ratio depends on the dose. The swelling ratio increased with increasing dose.

But as shown in Fig.10 and Fig.11, a poor correlation with the swelling ratio was observed for the dose and the dose rate in this study since the number of the data points is limited and irradiation conditions other than the dose varies at the same time. Furthermore, as a result of comparing with the literature data, as shown in Fig.12, the good correlation with the irradiation temperature was obtained, and the swelling ratio also showed a tendency to increase markedly from around 320°C. On the other hand, as shown in Fig.13, the clear relation between the swelling ratio and the dose was not observed.

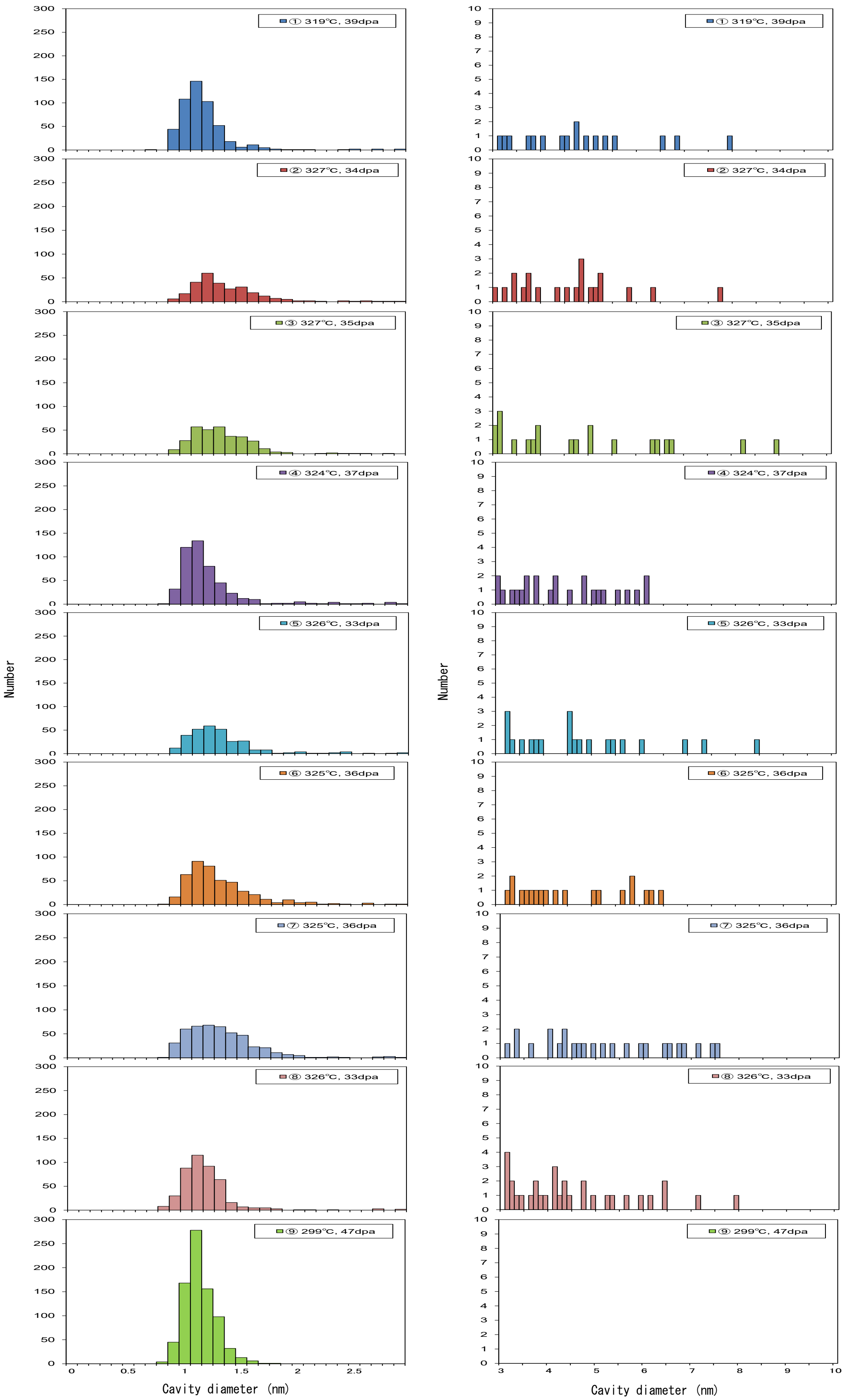


SW: Swelling ratio

Fig.7 Results of cavity observation under-focus TEM images

Table 2 Summary of swelling measurements

No.	Dose (dpa)	Irrad. temp. (°C)	Number of cavities (-)	Ave. diameter of cavities (nm)	Max. diameter of cavities (nm)	Density of cavities ($x10^{22}/m^3$)	Swelling ratio (%)
①	39	319	523	1.2	7.8	13.3	0.041
②	34	327	298	1.5	7.6	13.7	0.074
③	35	327	348	1.5	8.8	13.1	0.080
④	37	324	507	1.3	6.0	15.6	0.051
⑤	33	326	323	1.5	8.3	12.8	0.074
⑥	36	325	461	1.3	6.4	14.7	0.052
⑦	36	325	493	1.5	7.5	13.2	0.073
⑧	33	326	473	1.3	7.9	11.8	0.056
⑨	47	299	802	1.1	1.8	21.4	0.015



(a) He bubbles
(Cavity diameter ≦ 3nm)

(b) Voids
(Cavity diameter > 3nm)

Fig.8 Results of cavity size distribution for (a) He babbles and (b) voids

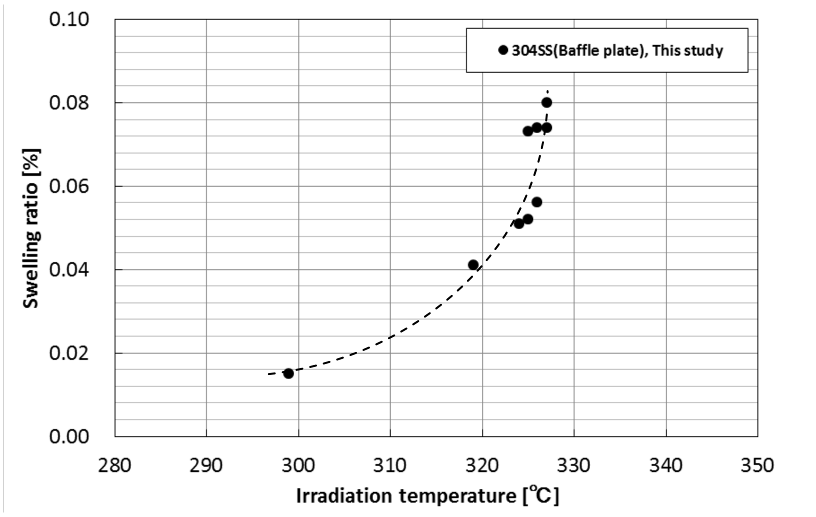


Fig.9 Correlation between swelling ratio and irradiation temperature

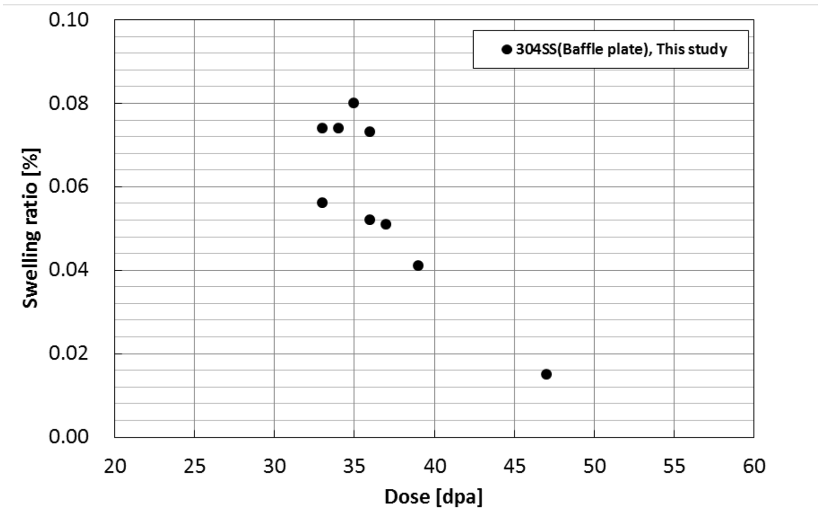


Fig.10 Correlation between swelling ratio and dose

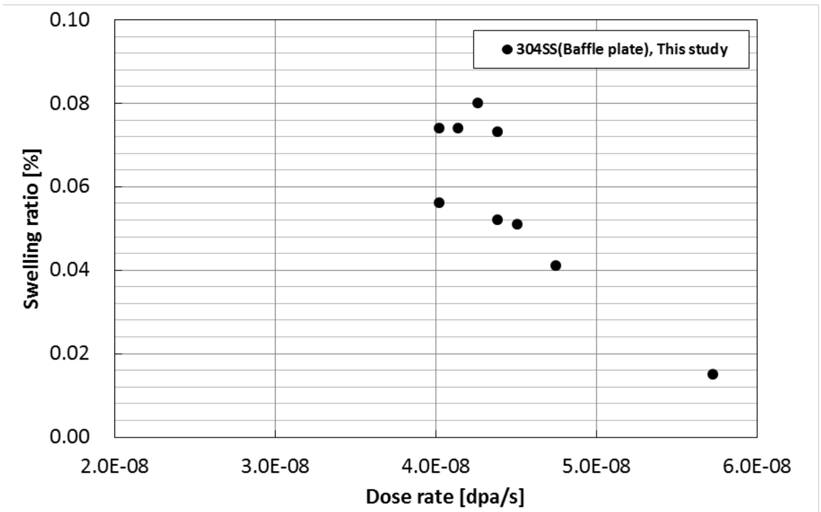


Fig.11 Correlation between swelling ratio and dose rate

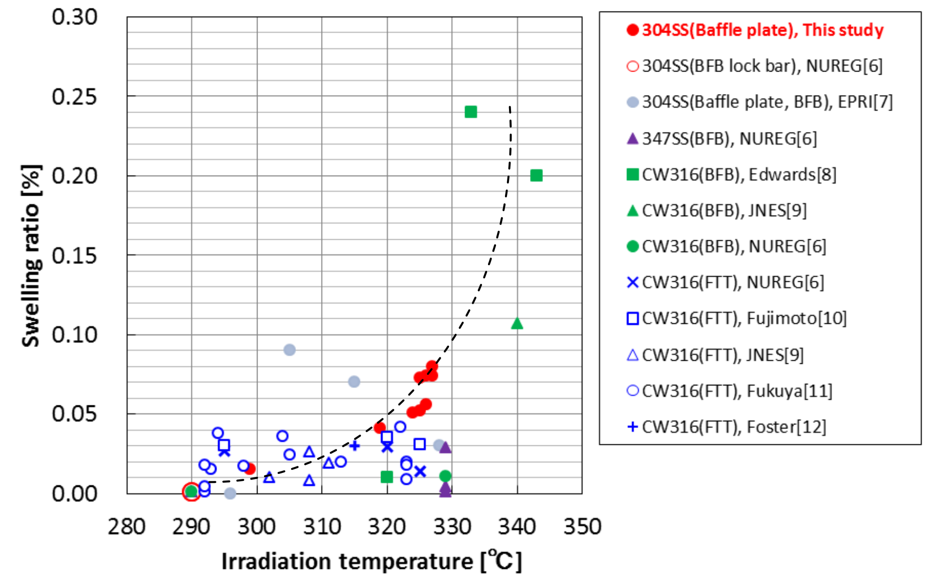


Fig.12 Comparison with literature swelling data as function of irradiation temperature for stainless steels irradiated in PWR

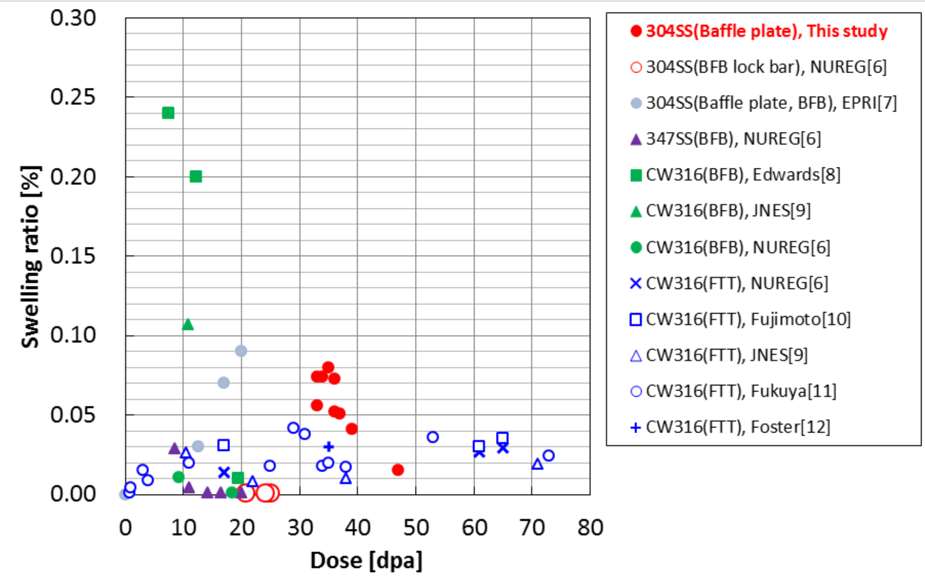


Fig.13 Comparison with literature swelling data as function of dose for stainless steels irradiated in PWR

(2) Comparison with Swelling Evaluation Equation
A comparison between the swelling measurement data in this study and the calculated by the swelling evaluation equation (modified dpa rate corrected Foster-Flinn equation [2, 3]) shown in the equation (2) is shown in Fig.14.

The swelling measurement data acquired in this study was slightly larger than the swelling evaluation equation, but good correlation was obtained. Furthermore, it was possible to obtain a result that well matches the rising temperature of the irradiation temperature in the swelling evaluation equation for the increasing trend of the swelling ratio in relation to the irradiation temperature.

In the PWR irradiation condition, remarkable swelling ratio was observed at around 320°C. It was possible to clarify the irradiation temperature dependence that transition from helium bubbles to voids is observed. Moreover, the reliability and validity of the swelling evaluation equation shown in the equation (2) could be obtained.

$$S = \Delta V / V_0 = \exp\left(-1.591 + 0.245T - 1.210T^2 - 1.384T^3 - 1.204T^4\right) \times \left(\phi / 1.25\right)^{-0.73} \times \left(F / 4.9\right)^2 \times 10^{-2} \quad \text{............ (2)}$$

$$T = \left(t - 490\right) / 100$$

Where,
S : swelling ratio [%]
t : irradiation temperature [°C]
ϕ : dose rate [dpa/s x 10^7]
F : dose [dpa]

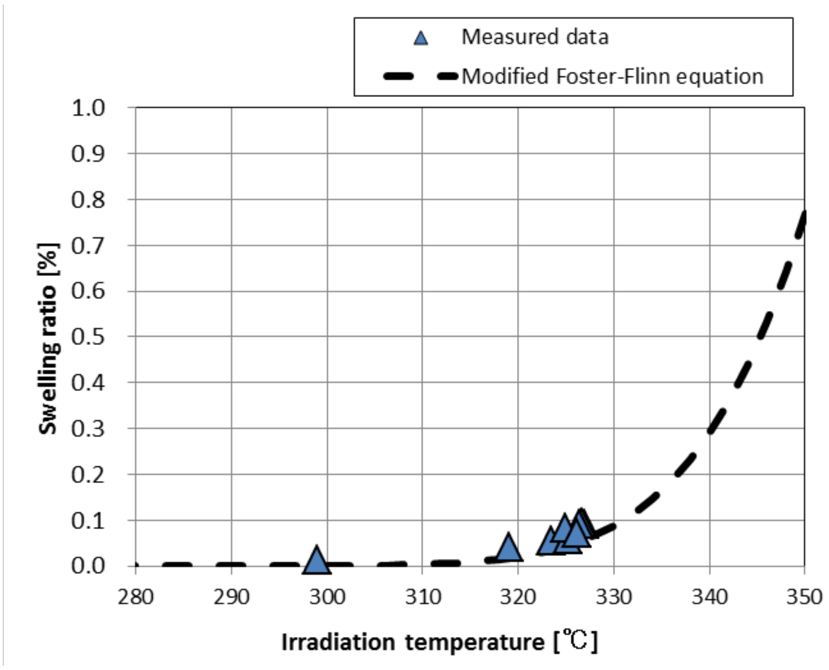


Fig.14 Comparison between measured data in this study and calculated data by modified dpa rate corrected Foster-Flinn equation for swelling ratio as function of the irradiation temperature

4. CONCLUSIONS

We investigated swelling characteristics of a coupon removed from a Type 304 stainless steel baffle plate of a decommissioned commercial PWR ZORITA plant, leading to the following results;

- ✓ The measured swelling ratio was 0.015%~0.080%, varying according to the dose (33dpa~47dpa) and irradiation temperature (299°C~327°C) at measurement points.
- ✓ Swelling ratios clearly increase with increasing irradiation temperature, while clear dependency of the swelling ratio for dose was not recognized as far as dose variation in this study.
- ✓ At lower irradiation temperature 320°C position, 1nm~3nm diameter helium bubbles were mainly observed, while at higher temperature (>320°C) positions, over 3nm diameter voids and less dense helium bubbles were observed. The growth from helium bubbles to voids seems to start from around irradiation temperature 320°C.
- ✓ The swelling evaluation equation called modified Foster-Flinn equation corresponds well with the obtained swelling data.
- ✓ In the future, we will try to improve the adequacy of the swelling evaluation equation for PWR by expanding irradiation material data at higher irradiation temperatures from baffle plates.

Acknowledgment

This study was done as a joint research program among Japanese PWR utilities and Mitsubishi Heavy Industries, LTD. (MHI). Grateful to EPRI Zorita Internals Research Project (ZIRP) for providing used coupon from Studsvik and its dose and irradiation temperature distribution calculations.

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