Improved technique to measure hydrogen concentration in the cross section of the fuel cladding

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Abstract

In the Reactor Fuel Examination Facility (RFEF) in JAEA, high temperature extraction method (HTE) has been used for hydrogen analysis. However, it has not been suitable technique in order to measure radial distribution of hydrogen concentration in the cross section of the fuel cladding.

Hydrogen concentration in the cross section of the fuel cladding can be measured with the backscattered electron image (BEI) analysis; In BEI on the cross section of the fuel cladding, brightness of zirconium hydride is different from that of matrix. BEI is processed to binary color between zirconium hydride and matrix to evaluate those areas. The area ratio of zirconium hydride to matrix is converted into hydrogen concentration.

This reports stated that condition on sample preparation, technique to take BEI such as accelerating voltage and sample current, and image processing procedures were improved for BEI method. In addition, the hydrogen concentration of un-irradiated cladding was measured with the improved BEI method to compare to the HTE. In the results, the hydrogen concentration by improved BEI method corresponded to hydrogen concentration by HTE method.

KEYWORDS: Hydrogen Concentration Measurement, Backscattered Electron Image, Fuel Cladding, High burn-up, Post Irradiation Examination

1. INTRODUCTION

For high burn-up fuel, increasing of hydrogen absorption to the fuel cladding is serious problem from the viewpoint of keeping the integrity of the fuel cladding. The hydrogen is absorbed to the fuel cladding with the formation of oxide layer during irradiation. The absorbed hydrogen that exceed solid solubility limit precipitates as the hydride phase. The hydride tends to precipitate it in the outer circumference side because hydrogen has a characteristic to migrate to the low temperature side, and high concentration of hydride causes the fuel cladding brittleness. Therefore it is important to evaluate hydrogen concentration at local area in the fuel cladding to confirm the safety margin.

The measurement technique for local area hydrogen concentration using backscattered electron image analysis (BEI method) [1] had been developed by Studsvik Nuclear AB, Sweden. The BEI method is very useful for the measurement of radial hydrogen concentration profile in the fuel cladding. In the RFEF, a sample preparation and image analysis procedures of the BEI method were improved to measure hydrogen concentration efficiently and precisely. In addition, confirmation tests were successfully carried out to apply that improved method for the Post-Irradiation Examinations (PIEs).

2. OUTLINE OF BEI METHOD

Hydrogen concentration is calculated using formula (1). Terms of right side of the formula are constant except for area fraction "F". The F is introduced by image processing of BEI obtained by SEM-EPMA.

$$Wt_H = Wt_\delta \times F \times \frac{\rho_\delta}{\rho_{Zr}(1 - F) + \rho_\delta \times F} \quad \cdots\cdots (1)$$

Wt_H = Hydrogen concentration (wt ppm)

Wt_δ = Hydrogen concentration in δ-phase hydride (17,570wt ppm)

F = Measured area fraction of hydride to matrix

ρ_δ = Density of δ-phase hydride (5.65g/cm^3)

ρ_{Zr} = Density of Zircaloy (6.54g/cm^3)

3. IMPROVEMENTS OF BEI METHOD

3.1 Polishing techniques for the observation plane

Procedure of sample preparation of conventional method such as metallography and SEM-EPMA is stated in the following.

- First step: The sample molded with epoxy resin is ground with emery paper from #400 to #4000.
- Second step: This sample is polished by diamond paste.

Seeing in the micro scale, the sample surface polished by diamond paste is not flat because polishing rate of Zircaloy matrix is faster than that of zirconium hydride due to difference in their hardness. To make the surface of the sample flat, third step is added.

- Third step: This sample is polished by the OPS which is silica alkaline suspension.

The OPS is mainly used to polish zirconium hydride.

3.2 Adjustment of SEM-EPMA

The SEM-EPMA and its' condition of taking BEI is stated in the following.

- SEM-EMPA with backscattered electron detector: JXA-8800 (manufactured by JEOL)
- Accelerating Voltage: 30keV (The higher accelerating voltage is, the higher the intensity of signal from back-scattered-electron is. But the higher accelerating voltage is, the lower depth resolution is.)
- Sample current: 5-6 $\times 10^{-8}$A (The higher sample current is, the higher the intensity of signal from back-scattered-electron is. But the higher sample current is, the lower the area resolution is.)
- Magnification of BEI: 800

3.3 Image processing technique for the hydride area fraction measurement

BEI is taken as some of the grayscale images which gray levels depend on the intensity of backscattered electron from the surface composition. The hydride is observed as black pixels on the BEI. In general image processing, image conversion from grayscale to binary image “black-and-white (B/W)” for distinction between hydride (black) and matrix (white) is performed according to a random threshold of the brightness. The area fraction of black (hydride) to white (matrix) pixels on the BEI is measured. However, there are blackish pixels rather than black pixels of the hydride in BEI, which are pixels of scratches, pits, etc. In addition, the pixels of the Zircaloy matrix have the brightness irregularity. If B/W conversion according to a brightness threshold is applied to BEI, all black and blackish pixels including scratches, pits and brightness irregularity are measured as the hydrides pixels. Therefore, it is important for accuracy measurement to distinguish the hydride pixels only from black and blackish pixels.

One of the image processing techniques, called “Color Gamut Selection (CGS) method”, is used to distinguish the hydride pixels only. Figure 1 shows schematic drawing of the CGS method. The CGS method has four steps,

1) Choose one “Criterion pixel“ on the center of a hydride region,

2) Select pixels which have about the same brightness as the “Criterion pixel”, and which are cascaded to the “Criterion pixel“.

3) After applying the CGS method to all hydride lines.

4) Remove background. (Remove the unselected pixels)

The selected pixels are estimate to hydride.

Figure 2 shows the comparison of B/W images converted by brightness threshold and CGS method. The CGS provides good distinction of the hydride pixels from black and blackish pixels, and the hydride area fraction is accurately measured by this method

4. CONFIRMATION TEST

In order to determine the reliability of the improved BEI method, the confirmation tests have been performed using the un-irradiated Zircaloy cladding tubes which have the different three types of hydrogen concentration. Each un-irradiated Zircaloy cladding tube has homogenous distribution of zirconium hydride. The hydrogen concentration measured by the improved BEI method is compared with that by high temperature extraction (HTE) method, which is good reliable method for the hydrogen concentration measurement.

BEI method is used to evaluate hydrogen concentration at local area and HTE method provides the average hydrogen concentration in the sample. Therefore, to compare these data between BEI and HTE method, the confirmation test was carried out with the following examination process.

[BEI method]

i. Take some of BEI from the outer to inner circumference continuously and combine those BEI

ii. Measure the hydride area fraction of combined BEI and calculate a hydrogen concentration

iii. Perform the same process on four directions, 0°, 90°, 180°, 270° and to calculate average hydrogen concentration of each direction

iv. The hydrogen concentration is determined with averaging those four measured data

[HTE method]

i. Divide a ring sliced sample into four pieces

ii. Measure the hydrogen concentration of each divided sample with hydrogen analyzer

iii. The hydrogen concentration is determined with averaging those four measured data

Figure 3 shows each hydrogen concentration by the BEI and HTE. These measured data show good agreement from 150ppm to 700ppm. It is said that the improved BEI method can provide the accurate hydrogen concentration at local area in the fuel cladding, that can't be measured by HTE method.

5. SUMMARY

In BEI method, the polishing techniques for the observation plane, adjustment of SEM-EPMA and the image processing technique for the hydride area fraction measurement were improved to measure local hydrogen concentration at RFEF. In the confirmation test with un-irradiated Zircaloy cladding, the hydrogen concentration measured by the improved BEI method gave good agreement with the results of HTE method from 150ppm to 700ppm. The improved BEI method is very effective to measure the radial hydrogen concentration profiles in the fuel cladding. As the next step, we will apply the improved BEI method to irradiated sample which has inhomogeneous distribution of zirconium hydride in the fuel cladding.

6. ACKNOWLEDGEMENTS

The HTE measurement and the operation of SEM-EPMA are supported by the Fuel Examination section. The samples are provided from the Fuel Safety Evaluation Research Group. The authors wish to express their sincere thanks to all of them.

7. REFERENCES

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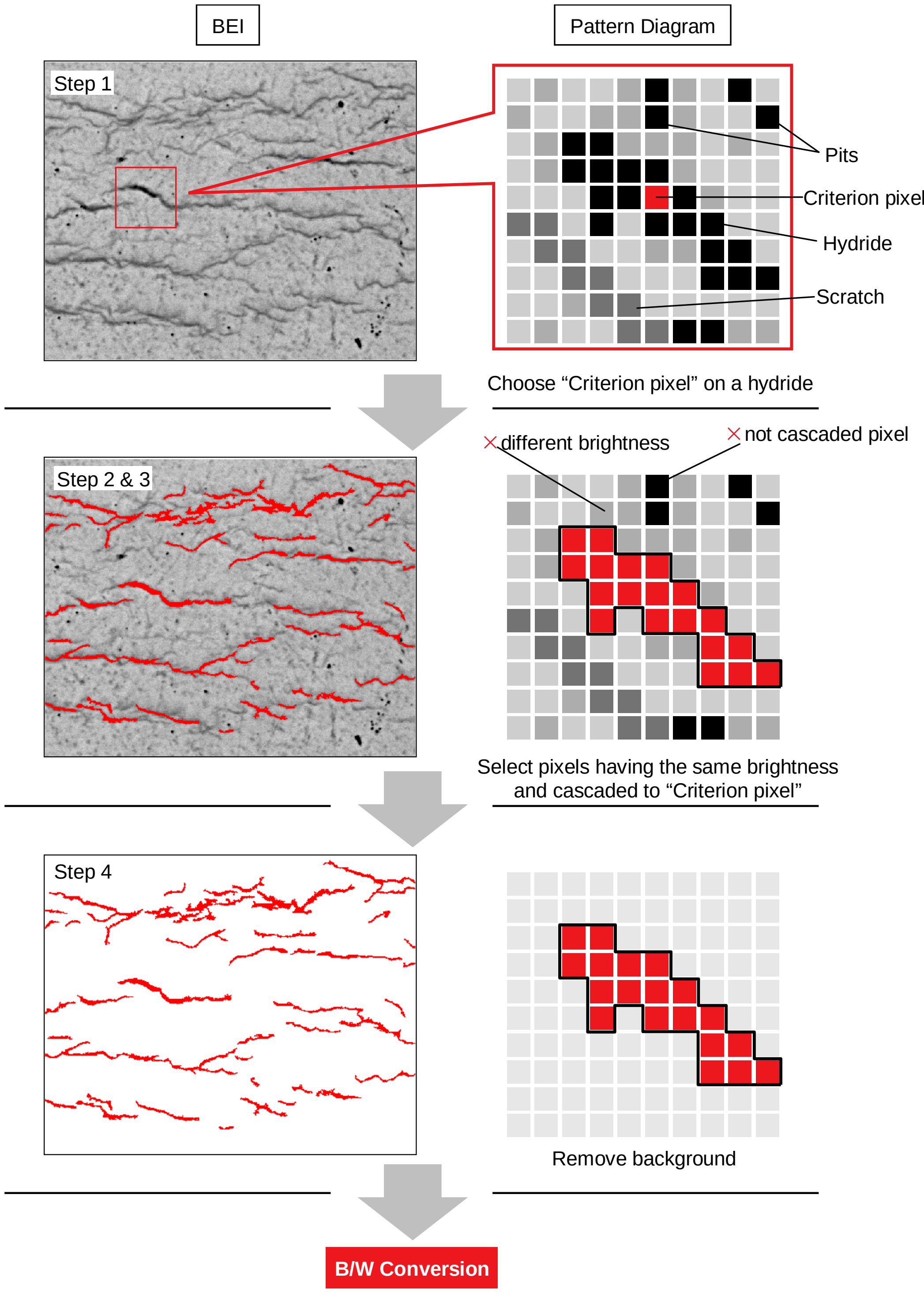

Figure 1 Schematic drawing of Color Gamut Selection Method

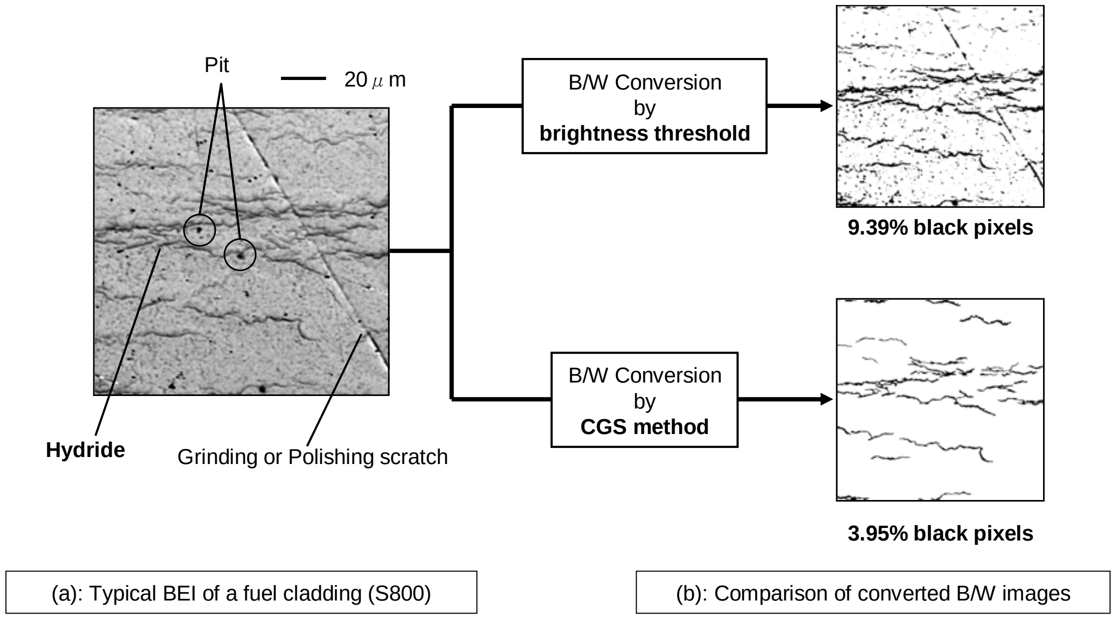

Figure 2 Typical BEI and B/W converted images

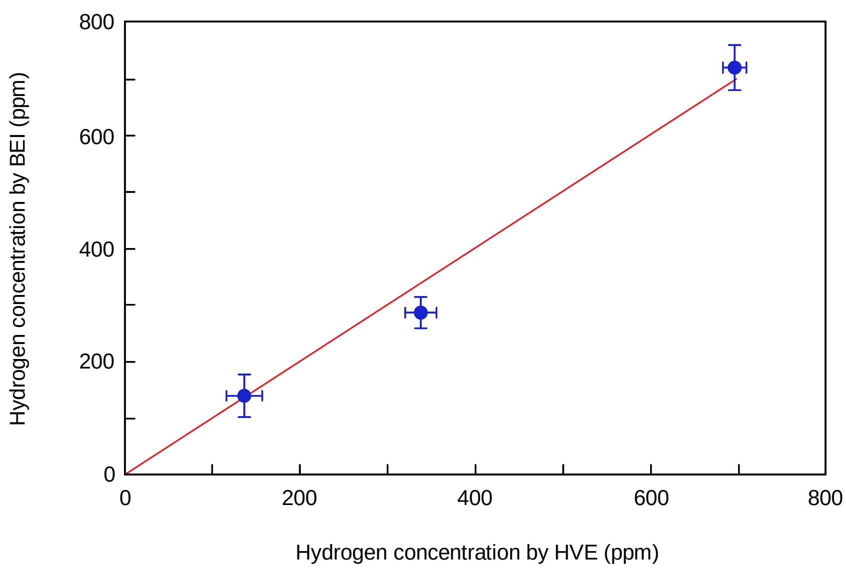

Figure 3 Result of confirmation test