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HYDRIDE REORIENTATION TEST FOR SPENT FUEL INTEGRITY

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ABSTRACT

Evaluation of spent fuel integrity for dry storage and transportation situation is getting more important because dry storage is significantly reconsidered for another extension beyond 200 years after Yucca mountain repository project was suspended in US. This very long term storage situation can cause more severe hydrogen related degradation as storage system temperature decreases.

Republic of Korea faces the PWR spent fuel pool saturation situation in reactor and dry storage like other countries seems to be most possible temporary solution in near future. Since spent fuels have to be transported to the final solution place after storage, possible spent fuel integrity degradation mechanisms must be thoroughly evaluated. Among them hydride reorientation(HR) has the highest priority deeply related to structural integrity during the transportation. Therefore, HR research has begun with testing system installation in hotcell area in Korea. The purpose of this testing are to follow the previous research works and compare our data with their result range, to suggest an empirical modeling including various parameter like hoop stress, temperature history (cooling time), and hydrogen contents (burnup).

Testing methods consist of 5 mm height ring type and 25 cm long tube type. Each method has advantages and disadvantages to analyze HR phenomena. Unirradiated cladding charged with hydrogen and irradiated cladding from spent fuel were used as testing specimen. Since Zircaloy-4 cladding occupies 78% of Korean spent fuel, this cladding is the main target material. ZIRLO, however, was also tested in the ring shape. This paper written at beginning stage of testing does not suggest any analysis opinion, but introduces current research situation and direction about hydride reorientation testing in Korea.

INTRODUCTION

During the normal operation in nuclear reactor, fuel cladding which is usually zirconium based alloy cannot avoid corrosion phenomena with high temperature water. Among the hydrogen amount separated from oxygen by corrosion, about 16% of hydrogen permeate into fuel cladding matrix and form a mixed precipitation with Zr, that is, hydrides when fuel temperature decreased rapidly after reactor operation. It is well known that the hydride precipitation direction has significant influence on the cladding material properties. For this reason, during the pilgering process cladding texture is intentionally controlled to introduce circumferential hydride precipitation preferably at HCP(Hexagonal Close-Packed) basal plane. These circumferentially precipitated hydrides play a hindrance role against outward hoop stress caused by internal gas pressure inside of cladding, radial crack propagation if it occurs, and outward creep movement of Zr-matrix dislocation.

Spent fuel accumulation problem at each nuclear reactor units needs an efficient solution like dry storage. During the spent fuel dry storage implementation, vacuum drying is the most important procedure because spent fuel temperature is getting high as heat transfer ability inside the cask(canister) decreased with water evaporation. After long wet storage in reactor pool, this re-heated spent fuel cladding temperature make the already circumferentially precipitated hydrides be resolved into cladding matrix again. Since spent fuel dry storage is a passive cooling system, spent fuel cladding temperature decreases very slowly over the decades. Hydrogen solubility of cladding also decreases along the temperature drop and this cause hydride precipitation again, but this time the precipitation direction tends to be radial because cladding itself still is willing to resolve the hoop stress caused by internal gas pressure and there is enough time for this phenomenon to occur with extremely slow cooling rate during several decades storage. This direction change of hydride precipitation is called „hydride re-orientation". It is well known that hydride reorientation is affected by hoop stress of internal cladding, temperature history, hydrogen amount, cooling rate, inherent clad material properties, thermal cycling, and irradiation effect.

After long-term storage, it is expected that certain amount of radial hydrides may exist in the cladding wall as a result of decreasing hydrogen solubility even though internal gas pressure also decreases. Inherently, dry storage is a temporary interim solution of spent fuel management, that is, spent fuels have to be transported to a final management solution like a reprocessing plant or a deep repository. In transportation situation, radially precipitated hydrides can offer a dangerous opportunity of easy crack propagation because hydride itself is very brittle and vulnerable for crack initiating when vibration and cycling impact occur during the transportation.

The decreased mechanical properties of clad by embrittlement are very critical at transportation accident case. Sandia National Laboratory research[1] result shows that a longitudinal tearing case(Mode-III) among the possible accident mode is very weak for radially hydrided cladding. Compressive impact on embrittled cladding can induce a longitudinal breach and massive fission gas release into cask(canister). In conclusion, hydride reorientation research is very important to the spent fuel cladding integrity evaluation during the dry storage operation and long distance transportation.

Republic of Korea has also faced the spent fuel saturation problem at the nuclear reactor pool. The spent fuel dry storage implementation is inevitable to solve this problem because other management options like recycling(pyroprocessing) and final deep repository needs more decades time to the commercial scale. Based on this background, hydride reorientation research has been performed to confirm that the already well organized regulatory technical criteria in advanced countries are applicable to domestic spent fuel which is suspected for its final bad material condition because of world-best high capacity factor.

According to the literature survey, two kinds of testing method has been used: one is the cladding ring specimen test with uni-axial tensile stress, the other one is the cladding tube specimen test with uniform radial stress. But, those research activities are usually focused on one method and performed with unirradiated cladding because of lots of testing limitation. Therefore, this study tried to perform both testing method with unirradiated/irradiated cladding to complement those research achievement.

TEST MATRIX

Hydride reorientation tests are undergoing with Zircaloy-4(Zry-4) and ZIRLO for ring and tube type specimen. Table 1 shows the currently performed test matrix items. The circle mark means that some of originally planned tests are performed, that is, those test matrixes did not finished yet. The blank matrix items mean that they are waiting for test. Tube tests were delayed compared to original schedule because there happen some problem with the pellet drilling machine in hot cell. Irradiated tests also need more time and efforts to obtain a good specimen, to improve shielding ability during the test and finally to achieve the regulatory organization‟s approval for irradiated material testing.

Table 1. Brief HR test matrix

	Ring		Tube	
	Zry-4	ZIRLO	Zry-4	ZIRLO
Non-irradiated	○	○	○	
Irradiated		○		

RING HR TEST

Ring test is the method that the hoop stress against cladding inner wall is simulated by tensile movement of inner grip on the ring type short length specimen. Usually 2 half cylinder or 3 piece mandrel with central dog bone are used for tensile movement grip with notched ring specimen. This notch can reduce the divergence of hoop stress and strain, but induce stress concentration on notched area because there are inherently stress gradient across the ring wall according to its clock direction. However, this notched specimen effect is higher in plastic region than elastic region, that means this effect is negligible at lower stress condition and hydride reorientation testing is usually performed in a elastic behavior region, therefore, the ring test without notch is preferable to more simple testing (especially for irradiated specimen) and advantageous effective observation on stress gradient across cladding wall for a once testing. Therefore, this study has used the 2 half cylinder pieces without notch and figure 1 shows this 2 piece stress raiser and simple comparison before/after HR test results schematically.

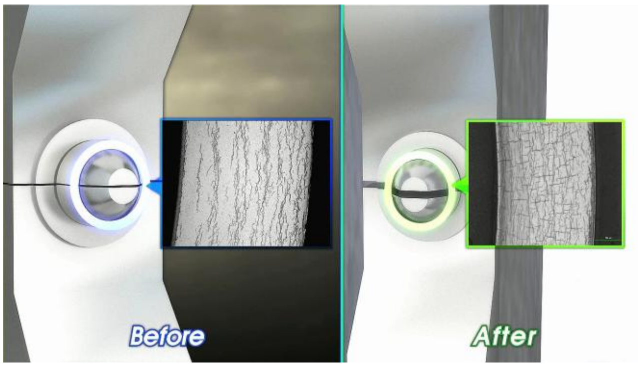


Fig 1. Schematical comparison before/after HR test results

There are some advantages of this method: relatively easy control of uniform temperature on short ring specimen, easy testing system installation compared to tube testing, small consumption of cladding, lots of various testing results at a given tube length, low radiation and simple shielding needs, and relatively easy handing of specimen.

Disadvantages of this testing method are following:

Essentially a finite element modeling(FEM) analysis is necessary to understand the stress gradient across the ring wall. Figure 2 shows that stress gradient across cladding wall(radial direction to the center of the ring) and stress distribution along the circumferential ring(clock direction) cause typical possible HR region where are outer side of 12 and 6 o‟clock, inner side of 3 and 9 o‟clock. Accurate FEM input material properties according to various temperature and stress are needed to make the FEM analysis results more reliable. This material data acquisition is very difficult for irradiated cladding specimen. Therefore, variation and low reliability of ring testing result are weaknesses compared to tube testing. Moreover, since hydride reorientation occurs locally at most high stress gradient region, it is difficult to get more additional material properties data using tensile or compression testing.

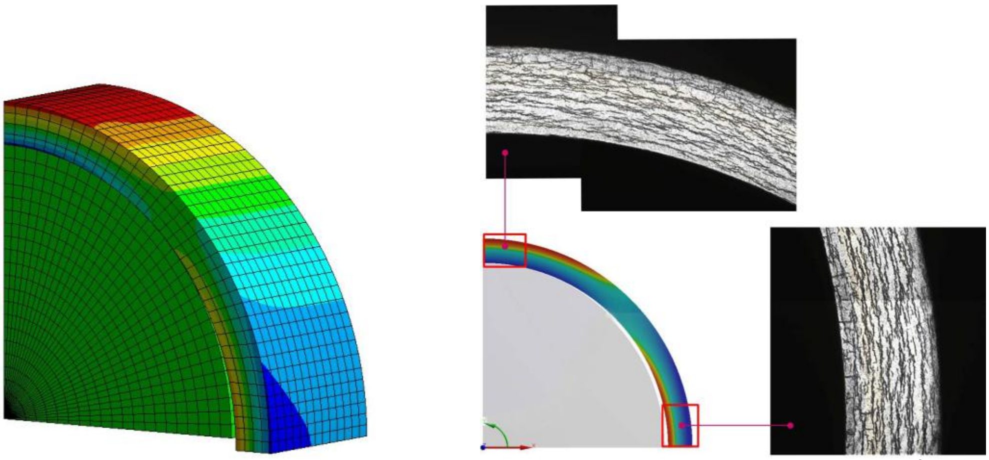


Fig 2. Finite element modeling(FEM) analysis about ring testing[2]

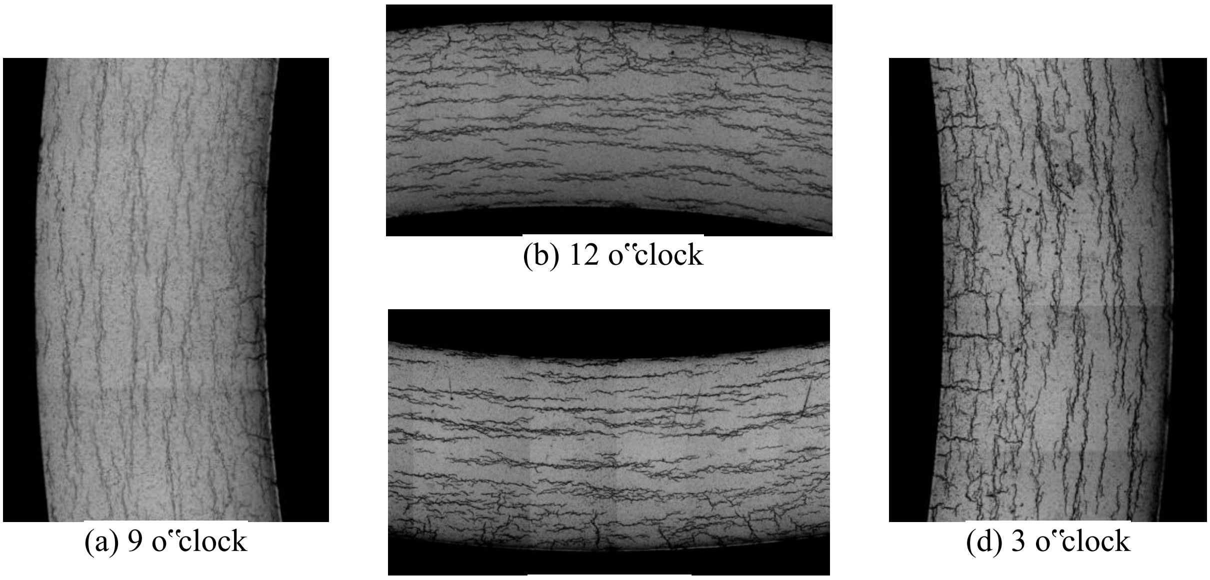
Unirradiated material

<Zry-4>

Gaseous hydrogen charging method in vacuum was used to simulate the unirradiated cladding as irradiated material. Thermal annealing treatment has been performed in order to make hydrogen distribution wide across the cladding wall cross-section.

Peak temperature range was 400~ 460℃, and cooling rate was unified with 0.5 ℃/min. Stress load was also unified with 123 N. Stress load, however, has been applied various temperature intervals to check differences between a case that constant stress load was applied for whole temperature cooling range from the peak to the room temperature value and a case that constant stress load was applied for only given 65 ℃ temperature intervals, for example, 400 ~ 335 ℃. The 65 ℃ temperature interval was based on the US regulatory guide for thermal cycling limitation[3]. Stepwised temperature interval testing is expected to give some information when HR occurs mainly. Cumulative effect, however, should be discussed together.

One of the unirradiated Zry-4 test result was shown in Figure 3. Peak temperature was 460 ℃, and stress load was applied during 400~335 ℃. After HR test, hydrogen analysis results indicated 360±10 ppm hydrogen concentration with this ring specimen. Each direction pictures shows good accordance with precedent FEM stress distribution. More detail analysis will be performed after obtaining enough testing results.



(b) 12 o‟clock

(a) 9 o‟clock

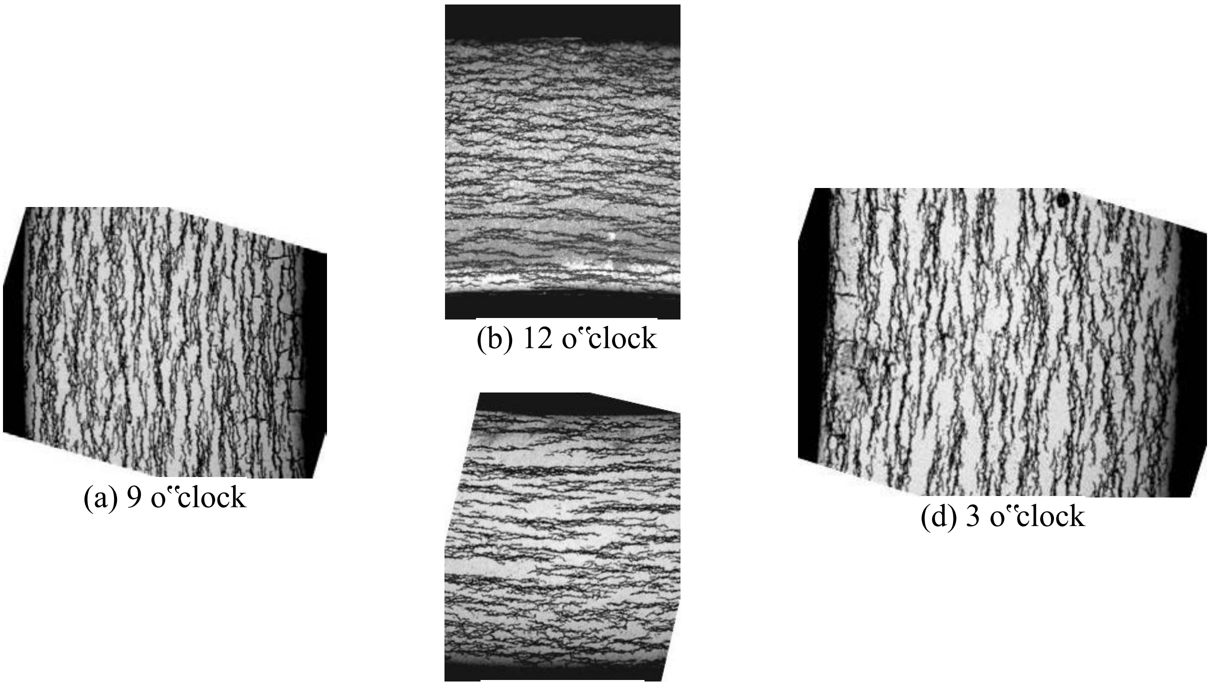
(d) 3 o‟clock

(c) 6 o‟clock

Fig 3. Optical microscope pictures for HR Ring Test result with unirradiated Zry-4

<ZIRLO>

One of the unirradiated ZIRLO test result was shown in Figure 4. Peak temperature was 400 ℃, and stress load was applied during 400~25 ℃. Hydrogen concentration analysis is not noticed yet, but its hydrogen concentration looks high enough compared to above Zry-4 specimen. Even though hydrogen concentration of ZIRLO is higher than Zry-4, the former‟s HR result is not clear than the latter‟s one. The number of testing is not pretty enough to introduce any kinds of clear conclusion about material differences, therefore additional tests with various condition change are necessary.



(b) 12 o‟clock

(a) 9 o‟clock

(d) 3 o‟clock

(c) 6 o‟clock

Fig 4. Optical microscope pictures for HR Ring Test result unirradiated ZIRLO

Irradiated material

<Zircaloy-4>
Irradiated Zry-4 cladding ring specimen supply is being delayed because of pellet drilling machine problem. As soon as this problem is solved, active test will be performed with this specimen.

<ZIRLO>
One of the irradiated ZIRLO test result was shown in Figure 5 with the original specimen picture before test. Axial position of this specimen is 30 cm below the upper plenum, and its local burnup was measured as around 57 GWd/MtU. Peak temperature was 400 ℃, and stress load was applied during 400~25 ℃. There are hydride rim region in the outer side of metal cladding very close to oxide film. While hydride reorientation occurrence is very clearly observed in the inner side of 3 and 9 o‟clock direction, it is not clear in the outer side of 12 and 6 o‟clock direction due to dense hydride concentration around this area. There are small sizes of hydride reorientation evidence in magnified pictures, but it does not seem to strongly threaten the cladding integrity because these hydrides are very short and connected crowdedly regardless of its direction in this region which disturb the continuous hydride-induced crack propagation.

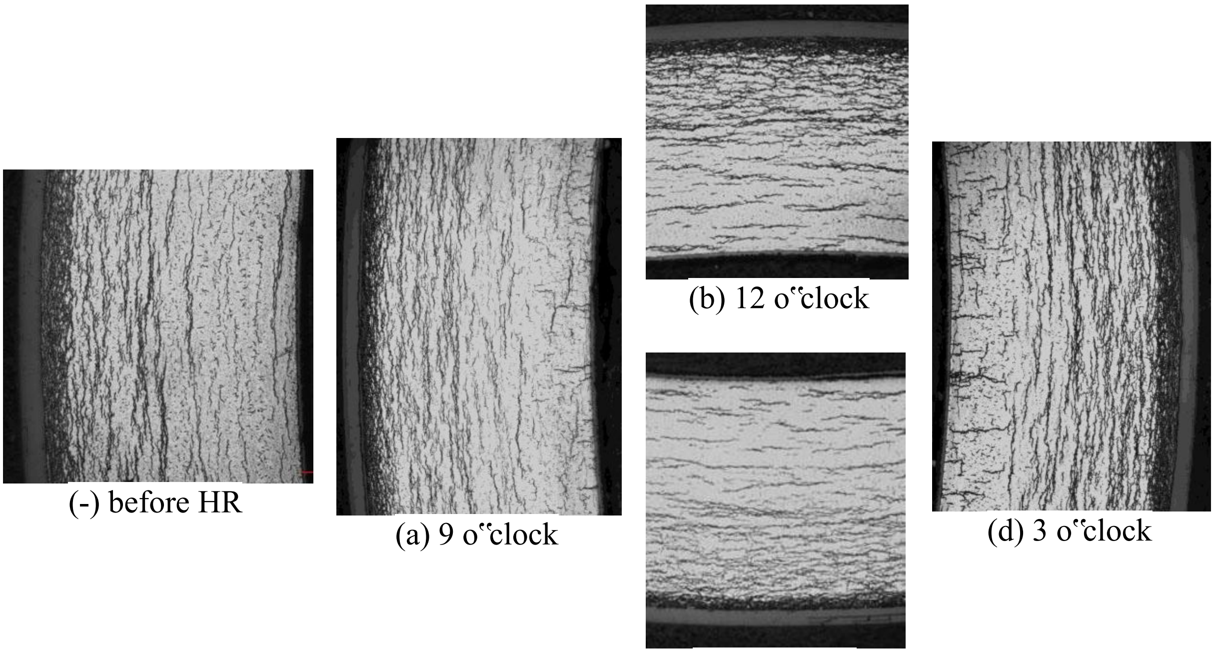


Fig 5. Optical microscope pictures for HR Ring Test result irradiated ZIRLO

TUBE HR TEST

In the tube test system, the top side of the tube is closed with high pressure fitting and the bottom side is connected to argon or helium noble gas supply line for internal gas pressure(hoop stress) simulation. Figure 6 helps to understand its system constitution. Since hydride reorientation testing needs slow cooling condition, it is necessary to compensate gas pressure drop according to temperature decrease in order to keep constant hoop stress inside the cladding tube if constant hoop stress is needed in some testing cases.

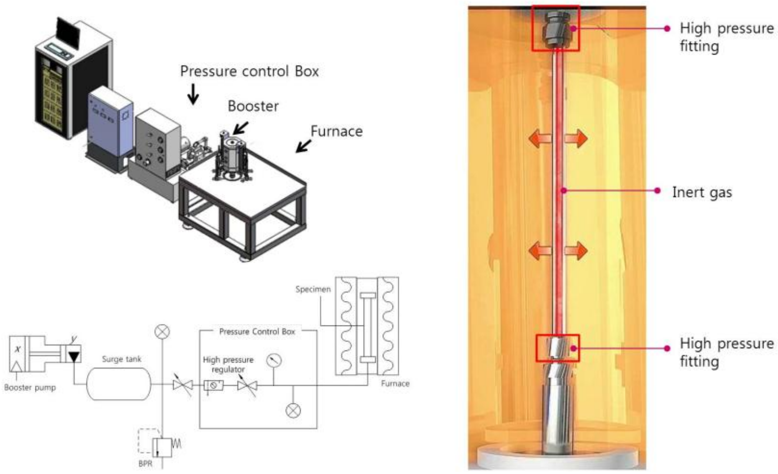


Fig 6. Tube test for hydride reorientation

Compared to ring testing method, there is no radial directivity and therefore, this method is more close to real spent fuel test simulation and it is easier to analyze the testing results. Moreover, it is possible to evaluate the changes of mechanical properties by performing a tensile and compression testing with some of the tube length after hydride reorientation tube experience. However, as far as the testing tube specimen is longer, there are some disadvantages: large consumption of tube (especially for irradiated specimen), difficulty of keeping uniform temperature distribution through the tube, difficulty of lengthy de-pellet drilling in hot cell, large furnace system to accommodate a long specimen, high radiation and massive shielding needs, and safety issue for accident cases because of high temperature and high pressurized gas testing.

As mentioned in test matrix section(Table 1), only unirradiated Zry-4 cladding tube test has been performed so far. Cladding tube was prepared 30 cm long at first step. In the next step, 25 cm length in the central region was used for test and the other upper & lower parts were used for „before test‟ optical microscope and hydrogen concentration analysis.

One testing case is representatively introduced in this paper among dozens of times cases. At the initial test stage, temperature was controlled to keep 400 ℃ and argon gas was supplied to impose 90 MPa hoop stress on cladding tube inner wall. After that, testing system was cooled down with 0.5 ℃/min rate which is same value with ring testing. At the final stage, testing was finished at room temperature, and the calculated hoop stress at that time was 73 MPa. In other words, this testing case did not use a constant temperature & hoop stress. Analysis for environmental conditions (constant or changing) needs more testing data. Hydrogen concentrations were 310~ 350 ppm range at axial points(H-1, H-2, H-3). Hydride morphologies as testing results are shown in Figure 7 where some hydride reorientations are observed evenly across the cladding wall. Four pictures show similar patterns because temperature was kept uniform(±1℃ in 15 cm central region) through the tube at a given time frame, hydrogen concentration were similar values along the tube, and finally the internal hoop stress was applied uniformly through the tube. Since this paper was written at beginning stage of whole testing schedule, analysis for the tube test results and correlation with ring test results was not prepared at confident level yet.

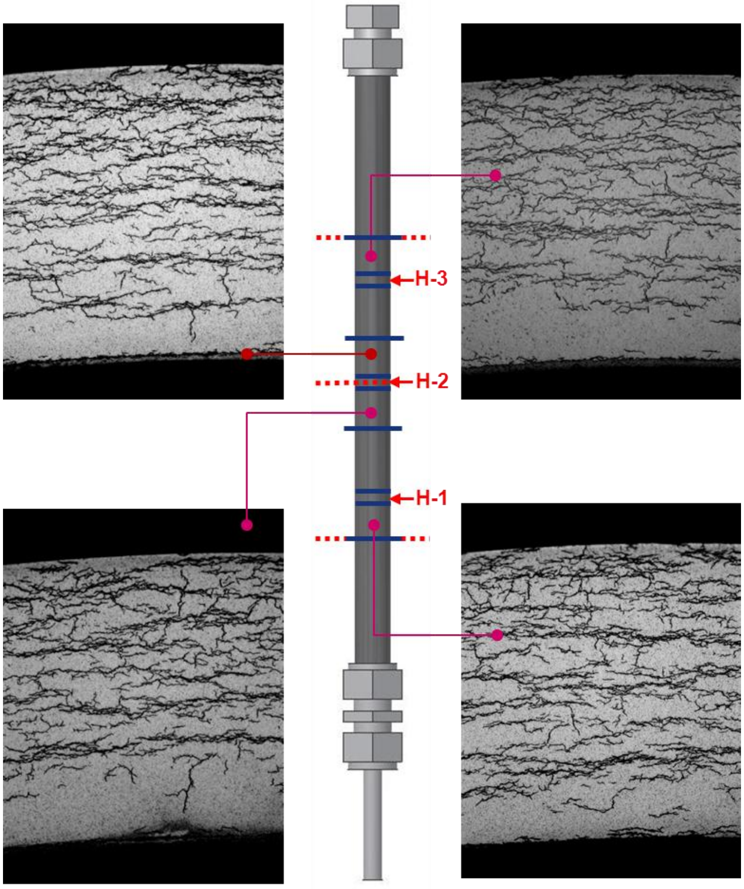


Fig 7. Hydride morphologies of unirradiated Zry-4 tube test[4]

PATH FORWARD

There are several work items remaining challengingly in this research like below:

- thermal cycled effect on HR
- tube test with simulated pellet inside
- HR test with hydride rim simulated specimen
- compression test for the rings produced from the post-tube test
- modeling for HR
- extended test for new cladding materials

CONCLUSION

Korea has faced the spent fuel dry storage era earnestly. Hydride reorientation is very important spent fuel cladding degradation mechanism in dry storage and transportation. Therefore, hydride reorientation research is underway at beginning stage in Korea. Based on the previous research achievement, ring & tube type, unirradiated & irradiated specimen tests are on-going together and their testing data & analysis results will be available in near future. Those are expected to contribute to the domestic regulatory criteria establishment and to the dry storage/transportation system applications.

ACKNOWLEDGMENTS

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