Post-Irradiation Examinations of SINQ Target-11

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Abstract. SINQ Target-11 was in operation in 2015 and 2016. It was shut down due to an incident in June 2016, during which the pressure drop in the cooling water loop increased significantly, and meanwhile, the activity of the cooling water also increased tremendously. The evidence indicated a serious failure of the target. As this is the first severe failure since SINQ came into operation in 1997, some post-irradiation examinations (PIE) were performed to analyze the failure mechanisms.

1. Introduction

The Swiss Spallation Neutron Source (SINQ) has been operated since 1997. Normally each target is operated for two years, which is mainly due to safety considerations as the structural materials receive a rather high irradiation dose and their mechanical properties decrease significantly. Although few small failures were observed in some targets and STIP specimen rods [1, 2], no serious failure was detected until the first one detected in Target-11 which was operated in 2015 and 2016. Target-11 was shut down on June 25, 2016, because the pressure drop in the cooling water loop increased nearly 100%. Furthermore, the activity of the cooling water had already increased by a factor of 3 since June 23. The evidence indicated a serious failure of the target.

The target was opened in a hot-cell (ATEC) next to the SINQ target station in June 2017. After removing the outer safety container (made of AlMg₃), it was observed that a large amount of lead (Pb), the target material contained in Zircaloy 2 cladding tubes, was melted and leaked out from the bottom of the target-block. Some empty tubes in the first row of the target were broken. During extracting some target rods for PIE, it was found that the rods in the high proton and neutron flux zone (the central region of the lower part) could not be pulled out. This implies that the core of the target was broken and the Pb was melted, which froze the rods in this part. Finally, six pieces of rods and tubes were successfully removed from the target for PIE. The analyses include visual and neutron radiography inspections, hardness measurement, Electron Probe Microanalysis (EPMA), and ring compression test. However, the ring compression test has not been done yet.

2. Experimental Results

2.1. Visual inspection

Figure 1 presents the bottom view of the target block after removing the safety container. It can be seen that Pb leaked out from the bottom of the target block, which might be driven by gravity even though the cooling water flew upwards at about 2 m/s speed in the gaps between the target rods. The first row at the bottom are empty tubes. The 4 tubes in the central part were either broken or bent.

Initially it was planned to take the rods from the high proton and neutron flux zone for PIE. However, it was found that those rods were not possible to be pulled out with manipulators. Finally, 6 rods/tubes from the positions indicated in Fig. 2 were removed from the target. Except for Rod TG11_P36-1, the other rods/tubes were seriously broken or damaged. The original length of these rods/tubes is about 12 cm. Thus, the pieces are mostly less than 50% of original length. It should be

note that, as there is essentially no cross-section reduction, the fracture of these rods/tubes looks brittle.

Fig. 1. (Right) The view of the target block of SINQ Target-11 after removing the AlMg3 safety container.

Fig. 2. Left: a sketch showing the position of the rods/tubes taken for PIE; Right: pictures of the rods/tubes taken out of the target.

2.2. Neutron radiography inspection

The rods/tubes were inspected with neutron radiography technique before they were transferred to PSI's hot laboratory. The inspection revealed the presence of Pb in the tubes (TG11_P0-3 to P0-5) extracted from the first row, as shown in Fig. 3. It is surprising to see so much Pb in the tubes. This implies that there was a large opening in each of them, from which the Pb was pushed into the tube. The opening was in the upper-central part of a tube, because no big opening can be seen on the lower surface (Fig. 1).

Except for the complete rod, TG11_P36-1, in the other rods/tubes the Pb at fracture part showed dark contrast. Considering the porous structure of Pb in this part, the dark contrast might be induced by moisture of heavy water in pores.

Fig. 3. (Right) Neutron radiography pictures showing the rods and tubes taken out from Target 11.

2.3. EPMA analysis

Three of the six rods/tubes, TG11_P0-5, TG11_P1-3 and TG11_P19-4, were selected for EPMA analysis. Specimens were short segments of about 3 mm, which were cut next to the fracture surface. The Pb inside the Zircaloy tubes was pushed out before mechanical polishing to reduce the activity of the specimens. Based on the neutronic calculation of Target-9 [3], the irradiation doses of the 3 EPMA specimens (TG11_P0-5-2, TG11_P1-3-2 and TG11_P19-4-2) are 24, 18 and 9.5 dpa, respectively. In the following sections the results of each specimen will be described.

2.3.1. TG11_P0-5-2

EPMA observations were performed at several locations on the cross-section of the tube as indicated in Fig. 4. Unfortunately, it was not possible to identify the proton beam direction for this specimen. Nevertheless, this seems not important, because the temperature of the wall of an empty tube in the first row is low, $<70^{\circ}\text{C}$, during normal operation [4]. The features observed in the seven positions of the sample (Fig. 4) are similar. As an example, Fig. 5 depicts the microstructure observed in position “C” in Fig. 4. Micrographs a, b and c correspond to inner surface, middle and outer surface regions of the tube. On both inner and outer surfaces, a thin layer of zirconium-deuteride was observed, which can be attributed to the corrosion due to the cooling water (D_2O). Inside the tube, the main features are the uniformly distributed precipitates and few hydrides.

2.3.2. TG11_P1-3-2

TG11_P1-3 is a Pb-filled rod. Therefore, the proton beam direction can be determined from the appearance of the Pb inside the tube and the holes for the wire lock, as roughly indicated in Fig. 6. As compared to the previous sample TG11_P0-5-2, the precipitate structure is similar. But, the hydride structure is quite different. Firstly, the appearance of hydrides is not homogeneous over the whole cross section of the tube. In the part around the position “G”, dense hydride lines of 10-20 μm length can be found across the whole thickness of the wall as shown in Fig. 7. Whereas in the part around the crack, namely position “F”, hydride lines are dense in the surface layer and decrease in density with the depth. Out of these areas, the situation is similar to that of sample TG11_P0-5-2. Secondly, on the outer surface nearby the crack there are some micro-cracks of about 20 μm depth as shown in Fig. 8. The location of the crack suggests that the stress should have arisen from the expansion of Pb (during the temperature rising period of proton beam trips). Since this rod is about 2

cm away from the central line of the target (Fig. 2), the irradiation dose and temperature distribution are not symmetric to the vertical diameter of the rod. It is expected that the half tube with the crack was closer to the central line of the target and with higher irradiation dose and temperature.

Figure 9 presents the results of element mapping obtained from a small area nearby the crack. It shows that the precipitates are enriched with Cr, Fe, Ni, and O, which are contained in the material at levels of 0.05-0.2 wt%. The Pb shown in the figure is believed due to contamination during sample preparation.

Fig. 4. (Right) Inspection positions of sample TG11_P0-5-2. The dashed circles represent the cross-section of the tube.

Fig. 5. Microstructure observed in position “C” in Fig. 4. Graphs a-c are for inner surface, middle area and outer surface regions. An enlarged view of the marked areas in a-c is presented under each graph. The scale in Fig. b is the same for Fig. a, b and c.

Fig. 6. (Right) Inspection positions of sample TG11_P1-3-2.

Fig. 7. Microstructure observed at position “G” in Fig. 6.

Fig. 8. The outer surface around the crack.

Fig. 9. Element mapping in an area nearby the crack.

2.3.3. TG11_P19-4-2

The proton beam direction of this rod can be also determined from the appearance of Pb inside the tube and the holes for the wire lock. Similarly, the inspection was performed in several positions as shown in Fig. 10. The observations showed some different features as compared to the previous rods.

Firstly, in the top region, as can be seen in the figure on the right, some material is missing on the inner and outer surfaces. The element mapping results presented in Fig. 11 demonstrate clearly that on the surface, Zircaloy was melted and then mixed with lead. This indicates that this rod experienced extremely high temperature.

Secondly, an oxide layer of about 5 micron thickness was observed on the outer surface of this rod, as shown in Fig. 12. The oxide layer is composed of two layers, outer and inner layers which can be seen in the Zr map. Naturally, there are some lead oxides in the outer layer.

Thirdly, the hydride structure in the tube is different from the previous two specimens. Comparing Fig. 13 with Fig. 7, one can see that the hydrides in this sample are much thicker. It is interesting to note that the precipitate structure completely changed in this region. Instead of the particles as shown in Fig. 9, elements Cr, Fe and Ni largely segregate at grain boundaries as shown in Fig. 14. Such a tremendous change can only be attributed to high temperature experienced rather than to irradiation, because the irradiation level of this rod is much lower than that of rod TG11_P1-3 or TG11_P0-5.

Fig. 10. Left: Inspection positions of sample TG11_P19-4-2; Right: enlarged view of the graph from position "I".

Fig. 11. Element maps from the area indicated in Fig. 10.

Fig. 12. (Right) A micrograph and element maps showing oxide layer formed on the outer surface of rod TG11_P19-4.

Fig. 13. Micrographs showing hydride structure formed in the top region of the Zircaloy tube of rod TG11_P19-4.

Fig. 14. A micrograph and element maps showing the precipitation in the top region of the Zircaloy tube of rod TG11_P19-4.

2.3.4. Hardness measurement

After EPMA analysis hardness measurement was conducted on these three specimens. Similar to the EPMA observations, the hardness measurement was performed in 45° step in the circumferential direction and 0.1 mm step in the radial direction starting from the position about 0.1 mm from the outer surface. The data are plotted in Fig. 15. The hardness of reference unirradiated material was measured (with another machine) to be 215 ± 12 kgf/mm². Irradiation-induced hardening effect (increase in hardness) can be clearly seen in specimens TG11_P0-5-2 and TG11_P1-3-2. For specimen TG11_P19-3-2 the hardness is in the same range as that of the reference material, namely no hardening. The “no hardening” should be attributed to the high temperature experienced.

Fig. 15. Hardness measured from specimens TG11_P0-5-2, TG11_P1-3-2 and TG11_P19-4-2.

3. Discussion

The irradiation dose of the specimens was evaluated. The best estimation is based on the neutronic calculation of irradiation experiment performed in Target- 9 (or STIP-6) [3], assuming that the distribution profile of the accumulated proton fluence is about the same for the two targets. The approach gives the following dose values for the three EPMA specimens:

- TG11_P0-5-2 (about 2 cm from the central line of the target): ~25 dpa
- TG11_P1-3-2 (about 1.7 cm from the central line of the target): ~18 dpa
- TG11_P19-4-2 (about 3.4 cm from the central line of the target): ~9.5 dpa

It is also interesting to note that the estimated maximum irradiation dose of the Zircaloy-2 tubes in Target 11 is about 40 dpa with about 1200 appm helium and 6600 appm hydrogen, corresponding to the 7.6 Ah proton charge received by the target.

Due to some reasons, the planned ring compression tests have not been conducted yet. The exact status of the ductility of the Zircaloy tubes is not clear. Here, the only data that can be used to assess the mechanical behavior of the target rods are the tensile test results of some specimens of Zircaloy 2 irradiated in STIP-2 (the 2nd experiment of the SINQ Target Irradiation Program [5]) or Target-4. In STIP-2 some specimens were manufactured from a Zircaloy-2 plate provided by the same producer of the Zircaloy tube. Although the production route of the plate might be different from that of the tube, it is the best representative for the tube. Fig. 16 presents the tensile engineering stress-strain curves of four Zircaloy-2 specimens irradiated in STIP-2 in the dose range of 10 to 31 dpa and tested at room temperature. Although the tensile curves of the four irradiated specimens are slightly

different, the four specimens demonstrate essentially the same behavior: strong irradiation-induced hardening and embrittlement effects. Attention should be paid to the strong embrittlement effect which results in very low ductility of the material.

Hardness of the four tensile specimens was also measured from the grip section of the specimens. The results show that the increase of hardness varies from about 60 to 100 kgf/mm² with irradiation dose increasing from 8 to 33 dpa (dose at the grip section), as shown in Fig. 17. Compared to these specimens, the hardness increase of the three specimens of Target 11 (Fig. 15) is less, but still significant for specimens TG11_P0-5-2 and TG11_P1-3-2. The lower hardening observed in these specimens may be due to the difference of materials and/or the temperature effect. For the specimen TG11_P19_4-2 of about 9 dpa, the hardening should be induced by the irradiation, but was not detected. An explanation is perhaps that the high temperature experienced during the incident annealed out the hardening effect. However, it should be noted that a short annealing at lower temperatures does not reduce the hardening, may even increase hardening of irradiated Zircaloy materials [6]. A STIP-2 specimen which was annealed at 800°C for 1 hour did not show any reduction of hardness. Transmission electron microscopy has to be performed to understand the microstructural changes induced by irradiation and annealing, which is unfortunately not possible now.

Fig. 16. Tensile engineering stress-strain curves of Zircaloy-2 specimens irradiated in STIP-2. The irradiation dose and temperature are as indicated.

Fig. 17. Increase of hardness vs. irradiation dose for specimens of Target 11 and the Zircaloy-2 specimens irradiated in STIP-2.

It should be addressed that all the results obtained from the specimens of Target 11 indicate the status of these few rods *after the incident*, rather than that directly *before the incident*. Similar investigation is being conducted on some specimens extracted from Zircaloy tubes of Target 9, which will depict the irradiation effects in the Zircaloy tubes during normal operation. However, it is clear that, at such irradiation doses (30-40 dpa for the high irradiation flux zone), the Zircaloy tubes were strongly embrittled by irradiation and the tube material significantly lost its ductility. In this case, the cycling thermal stress induced by the beam trips [7] may result in cracking of the tubes.

Although the empty tubes in the first row were already brittle, they should not break in the normal operation condition, because the stress level in the tube wall is low. The failure of the tubes should be caused by the melting of the upper-middle part of these tubes, where the melted Pb dropped down from the rods above them and blocked the water gaps around them.

It should be mentioned that in all of ten SINQ targets operated in the past, just three target rods showed damage (cracking) after operation. Two of them were with SS 316L cladding tubes irradiated in Target 4, the other was with a Zircaloy cladding tube in Target 8. The failure of the two SS 316L tubes was expected associated with over focused proton beam, which also caused damage of some STIP specimens. Like Target 11, Target 8 was also without any STIP specimens. There was no over focused beam experience for the target. The target received about 12 Ah proton charge, which resulted in a maximum irradiation dose of about 60 dpa in Zircaloy. This means the Zircaloy tubes were already brittle. A crack initiated in the tube under thermal stress induced by beam trips is possible. For Target 11, the irradiation dose is lower. However, it experienced more beam trips due to the operation of the ultra-cold neutron source (UCN) [8]. Since the Zircaloy tubes already received a maximum dose of 30-40 dpa, the material was getting into the brittle regime, cracks could be formed and developed at sites like micro-sized surface flaws or precipitates or hydrides in the surface layer under the cycling thermal stress (>100 MPa on tube surface [7]) induced by beam trips. Last not least, the short period of over-focused beam detected on June 23, 2016, namely 2 days before the incident, is believed to be the main reason for a large and sudden damage of some rods, which induced the great increase of radioactivity detected from the cooling water [8].

Conclusions

Post-irradiation examinations have been conducted on the several target rods extracted from SINQ Target 11. Visual inspection was performed when the target was opened and neutron imaging inspection was conducted on some rods extracted from the target. EPMA and hardness analyses were

done on three representative rods, TG11_P0-5, TG11_P1-3 and TG11_P19-4, which were of the highest irradiation doses of the extracted rods. The conclusions drawn from the results are that:

1. In the central (high proton and neutron flux) zone of the target, the Zircaloy tubes experienced very high temperature during the incident, which resulted in the melting of the Zircaloy tubes in the zone.
2. Strong irradiation-induced hardening was detected in the specimen of TG11_P0-5, a tube in the first row. A comparison with Zircaloy 2 specimens irradiated in STIP 2 suggests that the tube material was getting brittle.
3. The effects of the irradiation-induced embrittlement and the cycling thermal stress induced by beam trips on the damage of the target should not be excluded, although the over-focused proton beam is likely to be the main cause of the failure.

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