The Status of Studies on Structural Materials under High Energy Proton and Neutron Mixed Spectrum

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Abstract. The R&D of spallation targets with either high power or high proton beam intensity has been stimulated by increasing demands for different applications such as neutron scattering science and nuclear waste transmutation devices. One of key issues in the R&D is the understanding of the behavior of structural materials in severe irradiation environments in spallation targets. A worldwide effort has been focused on studying the radiation damage effects of high-energy protons and spallation neutrons on different structural materials by conducting irradiation experiments in the targets of the Swiss spallation neutron source SINQ at the Paul Scherrer Institut. In the last few years, a large number of specimens irradiated in the SINQ targets were examined in several institutes in Europe, Japan and USA. A significant progress has been achieved in the basic understanding of the radiation effects and a large amount data have been obtained from the related post-irradiation examinations.

1. Introduction

After the European Spallation Neutron Source (ESS), American Spallation Neutron Source (SNS) and Japanese Spallation Neutron Source (JSNS) projects started in the first half of the 1990s, the understanding of the behaviors of materials in the environments of spallation targets became a topic of crucial importance. The experiences from the existing spallation targets such as at ISIS (at the Rutherford Appleton Laboratory) and LANSCE (Los Alamos Neutron Science Center) indicated that the properties of materials could be rapidly degraded by the intensive irradiation of high energy protons and spallation neutrons and possible corrosion and embrittlement effects of coolants (water or liquid metals), which could strongly limit the lifetime of a high power (~1MW) target [1-3]. To support the R&D of ESS, SNS and JSNS, and the APT (Accelerator Production of Tritium) program as well, a wide international collaboration was initialized and three major actions were taken: 1) to hold regularly international workshops on spallation materials technology (IWSMT); 2) to analyze the spent parts from ISIS and LANSCE; and 2) to perform irradiation experiments at LANSCE and SINQ (the Swiss Spallation Neutron Source). In 1996 IWSMT-1 was held in Oak Ridge [4] and meanwhile, the spent parts of LANSCE and ISIS were shipped to Forschungszentrum Jülich. In addition, an irradiation experiment at LANSCE was started and the SINQ Target Irradiation Program (STIP) was initialized. In the following years in the second-half of the 1990s, a considerable amount of results were achieved from the analyses of spent parts and LANSCE irradiation, which were reported at IWSMT-2 to -4 [5-7]. The first STIP irradiation (STIP-I) was also conducted [8]. The displacement dose of all the materials in these irradiations (including those from the spent parts) was up to about 12 dpa (in Fe or steels). Higher doses up to about 20 dpa were obtained in the following STIP irradiation experiments, STIP-II to –V, performed between 2000 and 2008.

It should be pointed out that the STIP irradiation program has received great interests from the R&D program of the Accelerator Driven System (ADS) for nuclear waste transmutation. The PIE (post-irradiation examination) of STIP has been an important part in the relevant materials research programs of the European 5\(^{th}\) to 7\(^{th}\) Framework Programs. Last not least,
STIP is one of key experiments supporting the development of the so-called MEGAPIE (MEGAwatt PIlot Experiment) target which was successfully operated at SINQ in 1996 [9,10]. In this paper, some details of the LANSCE and STIP irradiation experiments will be described, and a brief overview of the results obtained from irradiated austenitic and ferritic/martensitic (FM) steels will be given.

2. Irradiation experiments

LANSCE irradiation experiment was carried out during 1996 and 1997 [11,12]. The maximum dose was about 12 dpa in steels. More than 5000 specimens were of different types: tensile, mini-bend bars, CT (compact-tensile), TEM (the transmission electron microscopy) and from various materials such as austenitic steels (e.g. SS 316LN and SS304L), FM steels (T91, 9Cr2WTaNbV, F82H, etc.), Inconel-718, Al6061, and some pure metals. The specimens were placed in two main irradiation zones in front of and behind the W-target (producing neutrons for neutron scattering science) in the proton beam line. In the zone in front of the W-target, the specimens were mainly irradiated with 800 MeV protons. The maximum dose of specimens in this zone was about 12 dpa with helium produced at a rate of about 180 appm He/dpa [13]. In the other zone, specimens received both protons passing through the W-target and spallation neutrons from the W-target at much lower fluxes. Both displacement dose and helium concentration were lower for the specimens in this zone. The irradiation temperature was limited to 164°C for the APT application.

Specimens irradiated in STIP experiments were placed just inside the SINQ spallation targets [8,14], as shown in Figure 1. Therefore, the specimens received both high energy protons and spallation neutrons. As the specimens had to be enclosed in tubes with an inner diameter of about 9.5 mm, the specimens were all miniature types designed for accessing mechanical properties such as tensile, fatigue, fracture toughness, ductile-to-brittle transition temperature (DBTT) and microstructural changes of materials. Because each SINQ solid target was in use for two years, every STIP irradiation experiment also lasted for two years. During 1998 and 2008 five experiments were done. In between, in 2006, the MEGAPIE liquid lead-bismuth eutectic (LBE) target was irradiated without any test specimens. However, the structural materials, T91 and SS 316L, of the components in the spallation zone of the target will serve as precious specimens for studying the synergetic effects of irradiation and LBE corrosion/embrittlement.

More than 40 kinds of materials were irradiated in STIP, including Fe-, Ni-, Al-, Zr-, Mo-, and W-alloys and pure metals e.g. Ta. Some specimens were irradiated in contact with stagnant Hg, LBE and Pb. Table 1 presents the irradiation conditions in terms of maximum dose in steels and maximum irradiation temperature. It should be noted that the dose and temperature distribution profiles are similar to that of the incident proton beam, which was approximately 2-D Gaussian distribution. However, with the distance increasing from bottom to top or centre to edge, the contribution of neutrons to displacement damage dose increases. This also resulted in a simultaneous decrease in He-to-dpa ratio, as the helium was mostly produced by protons. The value of the He-to-dpa ratio varied between 40 to 85 appm He/dpa.
Figure 1. Left: a sketch shows the positions of the specimen rods in the lower part of the target. Right: a photo shows the target block consisted of a frame holding the target and STIP rods and thermocouple wires.

Table 1. Materials and irradiation conditions of STIP-I to –V.

<table>
<thead>
<tr>
<th>Materials</th>
<th>STIP-I</th>
<th>STIP-II</th>
<th>STIP-III</th>
<th>STIP-IV</th>
<th>STIP-V</th>
</tr>
</thead>
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<tr>
<td>Austenitic steels</td>
<td>≤ 12 dpa</td>
<td>≤ 20 dpa</td>
<td>≤ 20 dpa</td>
<td>≤ 20 dpa</td>
<td>20 dpa</td>
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<tr>
<td></td>
<td>≤ 400°C</td>
<td>≤ 400°C</td>
<td>≤ 400°C</td>
<td>&lt; 400°C</td>
<td>400°C</td>
</tr>
<tr>
<td>FM steels (FMS)</td>
<td>≤ 12 dpa</td>
<td>≤ 20 dpa</td>
<td>≤ 20 dpa</td>
<td>≤ 25 dpa</td>
<td>20 dpa</td>
</tr>
<tr>
<td></td>
<td>≤ 360°C</td>
<td>≤ 400°C</td>
<td>≤ 800°C</td>
<td>&lt; 600°C</td>
<td>400°C</td>
</tr>
<tr>
<td>FMS-ODS</td>
<td>≤ 20 dpa</td>
<td>≤ 20 dpa</td>
<td>≤ 25 dpa</td>
<td>20 dpa</td>
<td></td>
</tr>
<tr>
<td></td>
<td>≤ 400°C</td>
<td>≤ 800°C</td>
<td>&lt; 600°C(?)</td>
<td>600°C</td>
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<tr>
<td>Ni-alloy</td>
<td>≤ 12 dpa</td>
<td>≤ 20 dpa</td>
<td>≤ 20 dpa</td>
<td>≤ 25 dpa</td>
<td>20 dpa</td>
</tr>
<tr>
<td></td>
<td>≤ 400°C</td>
<td>≤ 400°C</td>
<td>≤ 800°C</td>
<td>&lt; 600°C(?)</td>
<td>600°C</td>
</tr>
</tbody>
</table>
3. Results of austenitic steels

Most of specimens of the LANSCE irradiation experiment have been investigated, while only a part of STIP specimens has been tested. Up-to-date, all the tests were performed at ≤ 500°C and mostly at ≤ 400°C.

For SS316L (or SS316LN) a large amount of tensile data were obtained for doses up to 12 dpa. At above 12 dpa only few data were available. Figures 2a and 2b show, as examples, the tensile curves of SS316LN tested at room temperature and at the irradiation temperatures [15]. At room temperature, it shows that with increasing irradiation dose from 3 dpa to 9.7 dpa, the yield stress (YS) and ultimate tensile strength (UTS) increased while the uniform and total elongations decreased. This indicates a general trend of increasing hardening and embrittlement with increasing dose in the low temperature regime. At elevated temperatures ranging from 100°C to 400°C, compared to the results of the room temperature tests, it can be seen that higher testing temperatures had an evident effect on the strength and ductility. For the unirradiated specimens, the strength and ductility decreased with increasing testing temperature. For the irradiated specimens, as the irradiation temperature was increasing with proton flux (or with irradiation dose), the specimens of higher doses were tested at higher temperatures. Similar to the case of unirradiated specimens, the strength and ductility of the irradiated specimens are significantly reduced. For example, the yield strength of the 9.7 dpa specimens decreased from about 970 MPa at 25°C to about 750 MPa at 300°C, and meanwhile the strain-to-necking (STN) decreased from 9.6% to 1%.

Similar results were obtained from STIP specimens tested by different partners [16,17], SS316L/SS304L specimens irradiated at LANSCE [18,19], and SS304L of the spent ISIS target [20]. Figure 3 shows the dose dependence of (a) yield strength (YS) and ultimate tensile strength (UTS) (b) strain-to-necking (STN) and total elongation (TE) of austenitic steels irradiated either at LANSCE or SINQ spallation targets at < 400°C and tested at room temperature. It can be seen that the data of different austenitic steels have a rather good agreement. At least in the present dose range, the total elongation remains at a level about 10%, although the uniform elongation or strain-to-necking falls below 5% in some cases.
There are not many fracture toughness data available, particularly at elevated temperatures. The LANSCE irradiation data indicate that the fracture toughness of SS316L/304L decreases to about 60 MPa\(\sqrt{m}\) at about 8 dpa dose level for small CT specimens irradiated and tested at \(\leq 160^\circ C\) [21]. A few data of STIP irradiation at higher temperatures and higher doses show a value of less than 30 MPa\(\sqrt{m}\) for SS316LN at about 19 dpa and irradiated/tested at 400°C [22].

So far only a few small bend-fatigue specimens were tested by the Japanese team at room temperature [23]. The tests were conducted at a frequency of 26 Hz. The results show that irradiation to about 9 dpa does not affect the fatigue lifetime of JPCA and SS316F. It is known that irradiation induced degradation of fatigue lifetime decreases with increasing frequency [24].

The microstructural changes in SS316LN and its electron-beam welds (EBW) were observed with the TEM technique at PSI [25,26]. In solution-annealed (SA) EC316LN and its welds irradiated to doses up to 11 dpa and at temperatures ranging from 70°C to 350 °C, TEM investigations show that the main features of irradiation damage are high-density small black dot defects and large Frank loops. The density and size of the small dot defects are independent of irradiation dose with a mean size of 1 to 2 nm and a density of about \(2\times10^{23}\) m\(^{-3}\). The density of Frank loops varies little with dose, while the size increases with dose. Small bubbles were observed in specimens irradiated to 10 dpa or higher at temperatures above about 300°C.
Figure 2. Tensile stress-strain curves of unirradiated and irradiated SS316LN specimens tested at (a) room temperature and (b) irradiation temperatures. The numbers in the label of a curve indicate: the irradiation dose (in dpa), helium content (in appm) and irradiation temperature range in (a) and the test temperature, irradiation dose (in dpa), helium content (in appm) and irradiation temperature range in (b).
Figure 3. Dose dependence of (a) yield strength (YS) and ultimate tensile strength (UTS) (b) strain-to-necking (STN) and total elongation (TE) of austenitic steels irradiated either at LANSCE or SINQ spallation targets. The data are from ref. [15-21].

4. Results of FM steels

As FM steels were selected for key components in the MEGAPIE and ADS spallation targets, intensive investigations have been performed in the related R&D programs.
Tensile tests have been performed on specimens of conventional FM steels T91, EM10, HT9, EP823, and reduced activation steels 9Cr2WVTa, F82H, Optifer, Optimax [21,27-34]. Figure 4 shows the tensile data of the steels irradiated either in LANSCE or in STIP to doses between 3 and 20 dpa at temperatures between 80 and 350°C and tested at room temperature. The data indicate a trend that irradiation hardening increases with dose to about 17 dpa and then starts to saturate. In most of cases, the YS values are very close to the UTS values due to prompt necking of the specimens after irradiation. On the other hand, the ductility decreases significantly, particularly the uniform elongation which drops to about 1% or less in the present dose range. A number of specimens of doses above 17 dpa almost completely lost their ductility with a total elongation of less than 2%.

The fracture of these specimens shows a mixed mode of inter-granular and cleavage fracture in most cases. In some cases, for example two EP823 specimens of 13 and 16 dpa and one F82H specimen of about 10 dpa broke in the elastic region [31,32]. Some specimens irradiated in STIP-I to about 12 dpa (the maximum dose of STIP-I) show unexpected large ductility, which is believed to be due to the annealing effects of a high temperature excursion [29,32].

Tensile tests have also been performed at temperatures up to 450°C. Compared to the specimens tested at room temperature, the specimens tested at higher temperatures show a reduction in strength. Nevertheless, the hardening and embrittlement effects of the irradiated specimens are still significant. Figure 5 presents the results of the specimens irradiated in a temperature range of 200 – 300°C and tested at 250°C. It can be seen that, except for a reduction in strength of about 200 MPa, the results show similar features as the results of tests at room temperature shown in Figure 4.

The ductile-to-brittle transition temperature (DBTT) is an important parameter for FM steels, because the DBTT may greatly increase after irradiation. It is well known that the shift of DBTT (ΔDBTT) of different FM steels saturates at a dose level of 1-5 dpa after neutron irradiation [35-39]. Furthermore, some experiments demonstrate that the DBTT shift of FM steels after irradiation may be greatly affected by the helium content produced during irradiation [36-38]. Therefore, the He effects on the DBTT shift of FM steels are of great concern in fusion and spallation materials programs.

Charpy specimens of KLST and 1/3 CVN types from different FM steels have been irradiated in STIP to doses up to about 17 dpa at temperature below 300°C [39,40]. Figure 6 presents all the ΔDBTT data obtained from the specimens irradiated in STIP-I to STIP-III. Some data obtained from disc small punch (SP) tests are also included. The SP data (ΔDBTTSP) are converted to Charpy impact data (ΔDBTTCVN) with: ΔDBTTCVN = 2.5×ΔDBTTSP [28,41]. For comparison, some data of neutron irradiated T91 and Eurofer 97 [38,42] are included as well. This figure demonstrates clearly that the ΔDBTT of the different FM steels increases more or less linearly with dose after irradiation in spallation environment. At dose of about 5 dpa, the trend of ΔDBTT deviates from that of neutron irradiation, which is attributed to most likely helium effects. The helium effects on the DBTT shift can more clearly be seen in Figure 6b which shows a trend of ΔDBTT proportional to the helium content in the specimens.

The significances of the results are such: (1) helium can cause a very large DBTT shift, even much higher than that induced by irradiation hardening effects; (2) the DBTT shift increases more or less linearly with helium concentration; and (3) due to the helium induced DBTT shift, the lifetime of FM steels in fusion reactors and spallation targets where high helium contents are produced can be very limited.
The fracture toughness of some FM steels irradiated at LANSCE and STIP has been analyzed, although the data are still very limited [21,43,44]. The results demonstrate that the fracture toughness decreases continuously with irradiation dose. At a temperature below the DBTT of the irradiated material, the fracture toughness value falls below 50 MPa√m and the specimen breaks in a brittle fracture mode. For example, both T91 and F82H at about 15 dpa showed a fracture toughness value ≤ 50 MPa√m for testing at 250°C.

The irradiation introduced significant changes in the microstructure of the FM steels [45-48]. Irradiation induced defects, namely defect clusters or dislocation loops, were observed in all the irradiated specimens. At Ti≤300°C, the main features are small defect clusters of 1-2 nm in size and small dislocation loops of a few nm in size. With increasing irradiation dose, the densities of defect clusters and loops increase and the size of loops increases as well. At Ti>300°C, the densities of defect clusters and loops were found decreasing with increasing irradiation temperature. High-density helium bubbles with an average size above 1 nm can be observed in specimens with <=500 appm He and irradiated at ≤ 180°C. With increasing irradiation dose and temperature, the size of bubbles increases. Compared to that observed in SS316L, the bubble size is larger and the density is lower in FM steels in similar irradiation conditions.

Although the microstructure of the irradiated FM steels has been fairly studied, the correlation between microstructure and mechanical properties has not yet well been established. Modeling and more detailed microstructural analyses are needed to deepen the understanding.
Figure 4. Tensile data of different FM steels irradiated in either STIP or LANSCE. The specimens were irradiated at temperatures between 80 and 350°C and tested at room temperature. The data are compiled from ref. [27-34].
Figure 5. Tensile data of different FM steels irradiated in STIP. The specimens were irradiated at temperatures between 200 and 300°C and tested at 250°C. The data are compiled from ref. [28-34].
Figure 6. DBTT shift as a function of (a) irradiation dose and (b) helium concentration for FM steels irradiated in STIP. The data of neutron irradiation are from [38,42].

5. Outlook of the relevant research activities in the near future

In the next few years, the main activities in this field may focus on the following items.
(1) Continue the PIE of STIP specimens.

As previously mentioned, so far only a part of the specimens irradiated in STIP-I to –III has been analyzed. STIP-IV specimens will be available for PIE soon, and STIP-V will be available for PIE in 2011. It is anticipated that a rather complete database for austenitic and martensitic steels will be established within 3-5 years, which should meet the basic needs of the spallation source community.

(2) Perform PIE of the MEGAPIE target.

MEGAPIE was the first liquid Pb-Bi eutectic (LBE) spallation target of 1 MW power level. The maximum irradiation dose in the window was 6.8 dpa. The components exposed in the spallation zone are unique for studying the synergistic effects of irradiation and LBE corrosion/embrittlement. The results of the PIE will be very important for the R&D of the future ADS systems. The PIE tasks will be done by a close collaboration of all the MEGAPIE partner institutes.

(3) Conduct new STIP irradiation experiments.

New STIP experiments are still needed essentially for two reasons. Firstly, for a complete materials database for any nuclear application, it is important to investigate the irradiation dose dependence and irradiation temperature dependence of the materials properties. Such a database is necessary, for example, to extrapolate data for applications at higher irradiation dose levels. Secondly, the behaviors of special solid target structures such as W+cladding at high irradiation dose levels are not known, which is crucial for developing high power solid spallation targets [49].

References


[22] Y. Dai et al. to be published.


