Material issues relating to high power spallation neutron sources

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Abstract. Innovative researches using neutrons are being performed at the Materials and Life Science Experimental Facility (MLF) at the Japan Proton Accelerator Research Complex (J-PARC), in which a mercury target system is installed for MW-class pulse spallation neutron sources. In order to produce neutrons by the spallation reaction, proton beams are injected into the mercury target. At the moment, when the intense proton beam hits the target, pressure waves are generated in mercury because of the abrupt heat deposition. The pressure waves interact with the target vessel, leading to negative pressure that may cause cavitation along the vessel wall, i.e. on the interface between liquid and solid metals. On the other hand, the structural materials are subjected to irradiation damage due to protons and neutrons, very high cycle fatigue damages and so-called “liquid metal embrittlement”. That is, the structural materials must be said to be exposed to the extremely severe environments. In the paper, research and development relating to the material issues in the high power spallation neutron sources that has been performed so far at J-PARC is summarized.

1. Introduction
Neutrons are used for the innovative research that brings about breakthroughs in scientific and engineering fields, i.e. fuel cell, hydrogen embrittlement, protein structure, medicine, etc.. High power pulsed-spallation neutron sources are being operated at the “Materials and Life Science Experimental Facility” (MLF) of the “Japan Proton Accelerator Research Complex” (J-PARC) in Japan, and at the “Spallation Neutron Source” (SNS) in the US, which are standing on the way to increase the power up to MW-class [1, 2]. In both facilities, liquid mercury was selected as a target material to produce pulsed-neutrons by spallation reaction because of its high-neutron yield and efficient self-cooling performance, as shown in Figure 1.

At the moment that the protons bombard the mercury, impulsive pressure waves are generated in mercury due to the thermal shock. This results in the process of the pressure wave propagation, which leads to the aggressive cavitation in the mercury that imposes damage on the solid wall of the target vessel [3-7]. The impulsive pressure waves lead to the negative pressure through the propagation in mercury that may cause cavitation. Localized impacts by micro-jets and/or shock waves that are caused by cavitation bubble collapse impose so-called “pitting damage” on the vessel wall. The cavitation phenomenon becomes a crucial obstacle to increasing the power in the mercury target for the pulsed spallation neutron source. In fact, many penetrating pits due to cavitation were found in the inner wall in the used target vessels in the SNS, even under relatively low power [8]. In addition, the
structural materials of the target vessels are suffering from damage arising from high intensive neutron and proton irradiation, very high cycle fatigue, liquid metal embrittlement (LME), etc. [9-11].

Through theoretical and experimental investigations, the relationship between pressure wave conditions, aggressiveness of mercury cavitation, and damage growth behaviours in some solid metals can be understood. In order to mitigate the cavitation damage, we have applied two kinds of approaches: hardening of the vessel-wall and softening of the mercury. The former is achieved by coatings and/or surface improvements: Kolsterising®, nitriding, etc. [12]. The latter is enabled by the injection of micro-bubbles into the mercury, anticipating that the thermal energy caused by proton bombardment would be transformed to kinetic energy in the form of bubble oscillation [13, 14]. As for the structural materials investigation, the work carried out so far includes the very high cycle fatigue tests, the LME tests under mercury, irradiation effect evaluation tests using triple ions and the nano-indentation technique.

In this paper, research and development into techniques for the damage protection of the structure of the mercury target vessel is summarized.

Figure 1. Mercury target vessel at the MLF/J-PARC.

Figure 2. Typical examples of pitting damage surface protection treatments.
2. Mitigation of damage arising from cavitation

2.1. Hardening of vessel structural material

The high-cycle impact tests using an innovative electromagnetic impact machine, MIMTM, were carried out to understand the dynamic properties of mercury and the damage growth behaviour under localized impact due to cavitation in mercury [11].

More than 20 kinds of surface improvements: coating, implantation, etc. were examined experimentally to investigate suitable surface layer hardening protection against the cavitation erosion. The surface treatment was performed on solution-annealed austenitic stainless steel 316ss, which is the material used for the mercury target vessel. Figure 2 shows typical examples of pitting damage of samples with various surface treatments after imposing $10^6$ impacts: these are optical images and the depth profiles are taken by a laser scanning microscope. The level of damage is highly dependent on the surface treatments, and the plasma nitriding treatment exhibits the most resistant surface against the pitting damage, nevertheless, the fatigue strength is degraded by the nitriding treatment, because of the critical property changes along the interface between improved surface and substrate. We developed, therefore, an innovative surface treatment consisting of plasma carburizing and nitriding, PCN, and the thickness of the improved surface was determined by the stress distribution along the interface between the substrate and the surface layer through impact analyses [15-17]. This treatment is expected to extend the duty period from $10^6$ pulses to $10^7$ pulses. This treatment was applied to the beam window of the first target vessel in the J-PARC.

2.2. Softening of target material; mercury

The injection of micro-bubbles into flowing mercury is expected to reduce the pressure waves and suppress the cavitation bubble growth [18-26]. The initial compressive pressure wave is reduced by the absorption of the thermal expansion of mercury due to the contraction of micro-bubbles, as shown in Figure 3 [18, 19]. The effect is expected at the region where the heat is deposited and at the onset of pressure wave propagation. During the wave propagation, the micro-bubbles reduce the amplitude of the compressive pressure waves through attenuation of the pressure waves, by thermal dissipation of kinetic energy and wave dispersion. These effects are highly dependent of the size and number of injected bubbles. In the mercury target, the anticipated bubble condition for mitigation is that the injected gas bubble size and void fraction are 50 μm and more than $10^{-4}$, respectively [19].

![Figure 3](image)

**Figure 3.** Softening mechanism of mercury against proton beam pressure waves;
(a) absorption of the thermal expansion of mercury by the contraction of microbubbles,
(b) attenuation of the pressure waves by the thermal dissipation of kinetic energy.

The two mechanisms provided a marked mitigation to the compressive pressure induced by the proton beam bombardment. In fact, the mitigation effect was recognized in the 2005 WNR on-beam test. The dynamic response of the mercury pipe wall was measured by using a laser Doppler
vibrometer. The amplitude of velocity, which corresponds to the macroscopic pressure response caused by the proton beam bombardment, was clearly reduced by bubbling as shown in Figure 4 [25].

![Figure 4](image1.png)  
**Figure 4.** Displacement velocities measured at WNR on-beam tests.

![Figure 5](image2.png)  
**Figure 5.** Depth profiles of displacement damage and implanted ion concentrations computed by SRIM code.

### 3. Irradiation damage and evaluation method for nano-scale zone

The indentation technique, for measuring the load-depth curve, is a very promising technology for the evaluation of mechanical properties, in particular for micro- and nano-meter scale zone of materials, including very thin layers. The novel technology uses the indentation technique combined with numerical calculation, and is developed in order to evaluate the mechanical properties quantitatively. The inverse analyses with the Kalman filter were carried out on the loading and unloading curves measured by the instrumented indentation machine with the hemispherical indenter [27, 28]. The technique is applicable not only to the quantitative evaluation of material constants in the micro- and nano-meter scale zones, including very thin surface layers, *i.e.* corroded layers, ion implanted layers, but also to the small specimen testing technique for irradiated materials.

The properties of the interface materials used as boundary against severe environments seem vary gradually with the distance from the surface. Material integrity might be influenced by the surface property degradation. Target vessel materials used in spallation neutron sources are exposed to proton and neutron irradiation and mercury immersion environments. In order to evaluate the surface degradation due to such environments, triple ion beam irradiation was carried out to the materials treated with surface hardening that are the candidate materials for a beam window in the target vessel. The mechanical properties of the irradiated gradient surface layer were evaluated by the indentation technique with inverse analysis, using multi-layer model [23].

In order to investigate the irradiation effect on the surface hardening treated 316ss, the candidate window material for the mercury target vessel, triple ion beam irradiation testing using H⁺, He⁺, Ni³⁺ was carried out to take account of the target spallation condition. Figure 5 shows the depth profiles of displacement damage and implanted ion concentrations, computed with SRIM code [29]. The changes of mechanical properties and microstructure due to irradiation were evaluated by the nano-indentation technique and TEM observation of the micro-specimen cut by FIB (Focused Ion Beam), as shown Figure 6 [23]. As a result, no significant change was observed on the surface hardening treated layers of 316ss. The surface hardening treatment was applicable to the target window materials to reduce the erosion damage.
Figure 6. Cross-sectional TEM bright field images of un-irradiated and irradiated regions of 316ss and nitrided specimens. A few dislocation lines were observed in the un-irradiated 316ss, and larger dislocation loops with higher number density were formed in the irradiated 316ss. The martensitic structures with high dislocation density were observed in the un-irradiated nitriding, and there was hardly any change for the irradiated nitriding. It is considered that the dislocations of extremely high density in the martensitic structures are due to the nitriding suppressing the nucleation of radiation induced defect clusters by acting as a strong sink for point defects.

4. Liquid metal embrittlement
The effects of mercury immersion on fatigue crack propagation were investigated through three-point bending fatigue testing of notched specimens [30]. From the fatigue tests, in the low cycle fatigue region, the fatigue strength was seen to be slightly degraded by mercury immersion. In addition, the rate of increase in the measured notch opening distances of failed specimens with number of cycles, in the mercury case, was slightly larger than that for testing in air, regardless of the imposed stress, as shown Figure 7.

Figure 7. Effect of mercury immersion on crack propagation rates.

Figure 8. Roughness measurements of the fractured surfaces.
Figure 7 shows crack length as a function of the number of loading cycles. The rate of increase in the crack length as a function of the number of loading cycles can be classified as follows: incubation period A; a period, B, where the crack propagates gently according to the loading cycles; a period, C, where the crack propagates steadily and at a greater rate than during B; and finally, the fracture, at end point D. Region A, which corresponds to the early stage of Region I as shown in Figure 8, showed no differences between the mercury and air cases. In Region B, which the transitional region from Region I to II in Figure 8, a stepped crack length change was observed in the air case, which may be caused by unstable crack propagation. In Region C, a brittle fracture surface, *i.e.* an inter-granular fracture surface, was observed in the mercury case, as shown in Figure 9.

![Figure 9](image-url)  
*Figure 9. SEM images of the fracture surfaces at around 2 mm from the notch tip.*

5. **Summary**  
Impulsive pressure waves are a crucial obstacle to realizing the adoption of the mercury target for high power spallation neutron sources. High-cycle impact tests using an innovative electromagnetic impact machine, MIMTM, were performed to understand the dynamic properties of mercury, and the damage growth behaviour under localized impact due to cavitation in mercury. The hardening of the vessel surface and the injection of micro-bubbles, to soften the mercury and reduce the pressure wave amplitude, are expected to be useful for mitigating the localized impact damage resulting from cavitation. Additionally, the structural materials damage caused by proton and neutron irradiation, very high cycle fatigue and the so-called “liquid metal embrittlement” have been investigated through nano-indentation tests on triple-ion-implanted surface and crack propagation tests under mercury immersion. More detailed information on the research and development will be acquired from the references. As for very high cycle fatigue not described here because of space limitation, the recent progress is seen in ref. [31].

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