5.3 Material Issues for Spallation Target by GeV Proton Irradiation

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The importance of material issues in intence pulsed spalltion sources is described in connection with R&D of the JAERI 5 MW source.

1. Introduction

Materials used for a neutron target and its surroundings in intense spallation neutron sources suffer from serious radiation damage due to GeV proton and associated neutron irradiation. Material issues are most important which determine not only the life of important components such as a neutron target but also take an important role in radiation safety. The proton beam power in the JAERI pulsed spallation source is about 5 MW (1.5 GeV, 3.33 mA) and the peak beam current density at the incident proton beam window of the target is estimated to be in a range of 50-80 $\mu A/cm^2$, assuming a parabolic or Moffet beam distributions in the horizontal and the vertical directions with an elliptic beam footprint of approximately 70 cm^2. Since a heavy liquid metal target is the only realistic one which can accept such a high beam power, we chose mercury as the first candidate of the target material. The target container will have lateral dimensions lager than the beam footprint, at least, by about 4 cm in the horizontal direction and 3 cm in the vertical direction and an active length of about 70 cm with a dome window (see the illustration in Fig. 1). The proton beam power per pulse (repetition rate: 50 Hz, pulse length: about 1 μs) reaches at 100 kJ which brings about an energy deposition of about 60 kJ per pulse in mercury . In such conditions a beam window is considered to receive a static stress of approximately 100 MPa and a dynamic stress larger than 200 MPa, due to the pressure wave caused by such an energy input in a very short time duration (about 1 μs), and a radiation damage of several tens dpa per year. An important feature of the radiation damage due to the proton irradiation is much higher hydrogen and helium production rates per dpa than the case of 14 MeV neutron irradiation. The available data for such radiation damage are very scarce and the accumulation of the data base is highly desired.

In the present paper the importance of material problems in R&D of an intense pulsed spallation neutron source is given with recent R&D activities of the JAERI spallation source and some of recent activities on material irradiation and testing in the world community are shortly reviewed.

2. Operational experience of spallation targets

Many uranium (U, depleted or enriched) targets have so far been used in various spallation neutron facilities of small and medium proton beam powers (KENS: 3-5 kW; INNS: 6-8 kW; ISIS: 160 kW), due to their higher neutron yield per proton compared to other non-actinide heavy metal targets such as tantalum (Ta), tungsten (W), etc.; about 1.7 times higher yield than a latter in case of a depleted U-target, about 4 times or more in case of a highly enriched one. However, the service life of those U-targets were much shorter than expected. For example, in the worst case at ISIS the shortest life was approximately only one month. Table 1 summarizes important operational records of various U-targets so far been used (total number of thermal cycles due to frequent proton-beam trips from the accelerator, total number of protons incident upon the target etc. by the end of their lives) together with some data on Ta- targets used at ISIS, based on the reported data [1-3]. In case of U-targets, it used to be considered as the end of the service life, when an appreciable, sometimes a detectable, amount of fission products appeared in the primary cooling water or in its cover gas. In case of a Ta target, it is still not clear when we shall stop the operation.

All the U-targets in the table were water (H_2O or D_2O) cooled solid targets, consisting of numbers of target discs of appropriate thicknesses to ensure the necessary heat removal. Each target disc was clad by Zircaloy-2 (diffusion bonding by HIP). In spite of various efforts so far been devoted to improve the service life, for example, adopting a much lower operating temperature than 400 ºC, above which the swelling becomes serious, a proper alloying, making fine grains in metal U, etc., it was not successful to overcome the situation up to now.

Table 1 Some important records of neutron targets so far been used at various spallation neutron facilities

Target	Proton Energy, MeV	Thermal Cycles	Total Protons, mA hrs	Peak Temperature, ˚C	Time-average beam current, μA	Relative Total Number of Fissions to U #5
ISIS U #1	780	—	92.4	—	30	0.31
ISIS U #2	800	40000	53.1	120	40	0.18
ISIS U #3	800	10389	174.9	130	65	0.59
ISIS U #4	800	4147	138.8	150	75	0.47
ISIS U #5	800	5074	295.6	165	90	1.00
ISIS U #6	800	2628	126.1	180	110	0.43
ISIS U #7	800	1805	107.2	215	125	0.36
ISIS U #8		Not Used			—	—
ISIS U #9	800	815	113.2		150	0.381
IPNS Depleted #1	450	89600	240.0	225	10 - 14	0.39
IPNS Enriched #1	450	28000	128.8	175	14 - 16	1.07
KENS U #1	500	~ 40000	~ 50	~ 120	~ 5	~ 0.1
ISIS Ta #1	800	73378	1751.6		170	3.80**
ISIS Ta #2*	800	21138	618.1		170	1.34**

* Still in use

** Relative value of total number of neutrons to ISIS U #5

Therefore, It is being commonly recognized that the use of an U-target beyond the proton beam power at present ISIS, 160 kW, would be very difficult and impractical. Thus, the use of a non-actinide target becomes unique solution at a higher power level. The ISIS Ta#1 target had been used up to about 1750 mA-hr in the integrated proton current, almost 6 times as high as the highest value of the U-target (ISIS U#5). This target retired, not due to a failure but just for destructive inspections. Although we have to wait the results of the tests on the Ta#1 discs (non-clad) for detailed information on radiation damage, it has been reported that the discs seemed to have an enough ductility yet, suggesting that such a solid target could be used up to a proton-beam power level of about 1 MW. However at 5 MW the above time-integrated current can be reached only by 3 weeks and when we consider other factors than radiation damage, the use of a liquid metal target would be more practical than a solid target.

3. Liquid metal target

Among the candidates of liquid metal targets (Mercury (Hg), lead bismuth eutectic (Pb-Bi), molten lead (Pb)), Hg has various advantage over other candidate due to various reasons; (1) no radiation damage in target material itself, (2) liquid state even at room temperature (no need of preheating), (3) higher atomic number density than other two candidates, resulting in a higher neutron luminosity from target, etc.

Figure1 shows the calculated axial distributions of leakage fast neutrons from Hg targets towards moderator, determined at 2 cm from the target surface, for a cylindrical and a flat (rectangular cross section) targets, compared with those of Pb-Bi ones. It will be clear from this figure that an Hg target is always superior to Pb-Bi one and a flat target, accordingly with a flat proton-beam footprint, is much better than a cylindrical one.

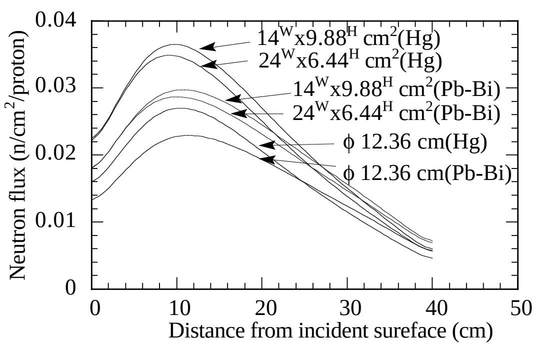

Fig. 1 Spatial (axial) distribution of leakage neutron intensities form various targets

4. JAERI 5 MW pulsed spallation neutron source

The JAERI 5 MW pulsed spallation source aims at realizing the highest slow neutron intensity worldwide, mainly for neutron scattering experiments for condensed matter research [4,5]. For this purpose R&D of a high-efficiency target-moderator-reflector system becomes indispensable. We have proposed an advanced concept for this as shown in Fig. 2 [6,7]. Important advantages of the present configuration are (1) the use of optimized flat target with flat proton beam, (2) a new target-moderator configuration which enables all the moderator to sit at the highest fast- neutron luminosity region on the target, (3) the use of coupled liquid (supercritical) hydrogen moderator with premoderator [8-11] for high-intensity and high-resolution cold neutron experiments, (4) a proposed new concept of target-moderator coupling "extended premoderator" for above mentioned moderators [12], etc. With extensive optimization studies based on the above concept of target-moderator-reflector system, following neutronic performances can be predicted;

(i) time averaged cold neutron intensity per MW of proton beam is approximately comparable to the one forth value of that from the second cold neutron source in the high-flux reactor at ILL, Grenoble, as shown in Fig. 3. This means that at 5 MW the projected source could provide 1.25 times higher time-averaged cold neutron intensity than the ILL. From the predicted pulse characteristics it turns out that the peak cold neutron intensity at 50 Hz is approximately 100 times higher than the ILL, with which many breakthroughs in various fields of research can be expected;.

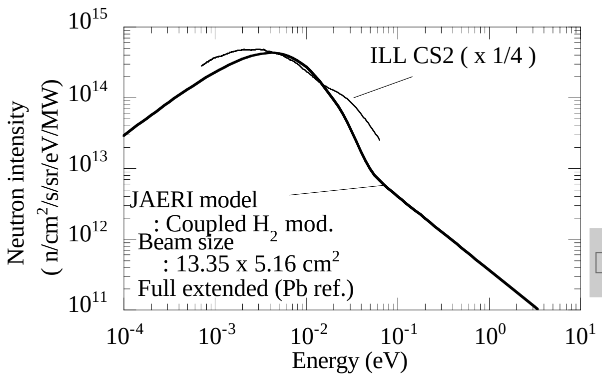

Fig. 3 Neutron spectral intensities from the moderator

(ii) It was confirmed that all moderators could be positioned approximately at the highesr luminosity region witout any cross talk between adjacenet moderators, resulting in the highest slow neutron intensities.

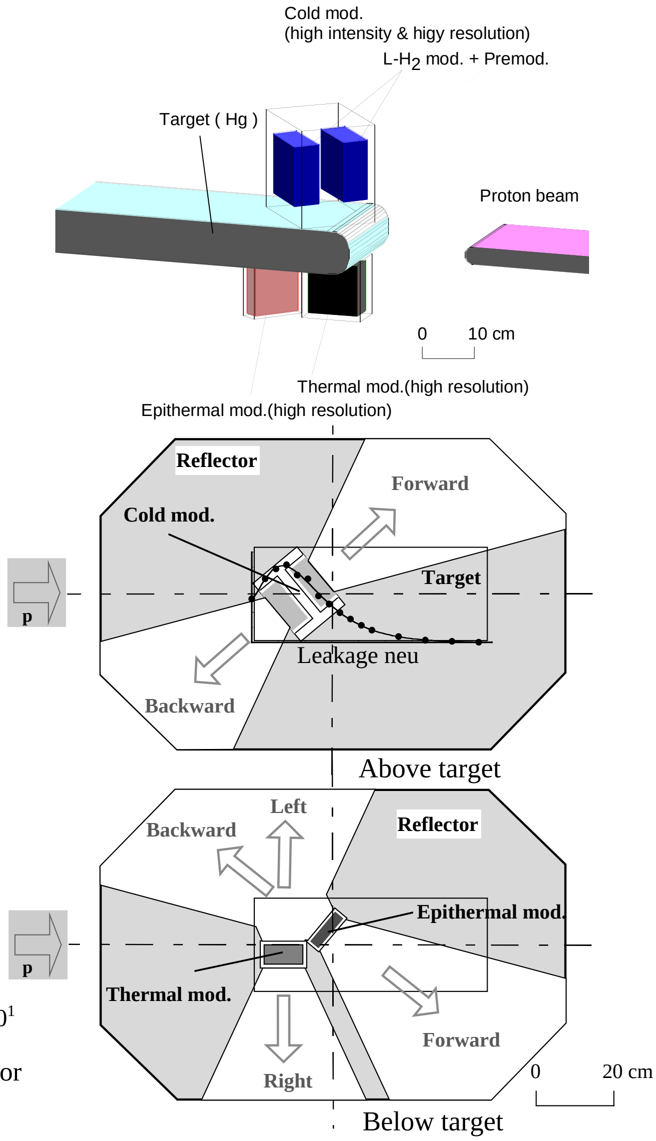

Fig. 2 Layout of target-moderator-reflector system

5. Engineering issues

In order to realize the above performance we have to solve various engineering problems. Figure 4 shows the energy deposition in an Hg target along the beam center line for 1.5 GeV proton incidence [13,14]. Here, the maximum proton beam current density was assumed to be 48 μA/cm^2, whatever the beam profile is. Note that the maximum power density in the target reaches at 1.75 kW/cm^3 (1.75 GW/m^3), which is much higher than a corresponding value in a power reactor and that this is the time-average

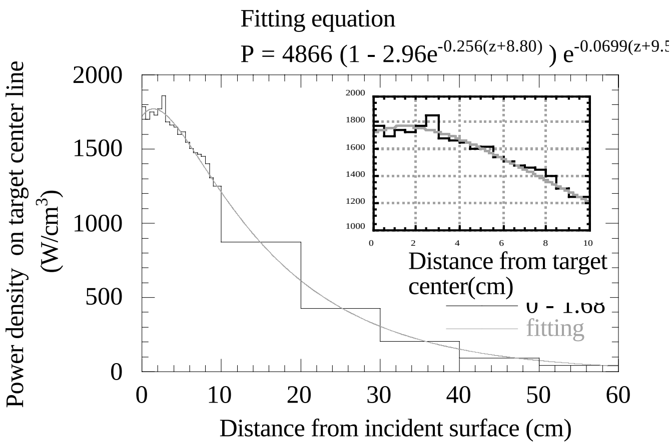

Fig. 4 Energy deposition in target as a function of distance from incident surface (cylindrical Hg target of 12.86 cm ϕ)

power densities, not at the pulse peak. The proton beam power of 5 MW corresponds to 100 kJ/pulse at 50 Hz which is given in a very short time of about 1 μs. Various important parameters and typical values for an Hg target, operated at a proton beam power of 5 MW with a repetition rate of 50 Hz. are summarized in Table 2.

Table 2 Main parameters of JAERI 5 MW Hg target

Parameter	JAERI SNS (expected values)	AGS Tests
Proton beam energy (GeV)	1.5	23
Protons per pulse	4.2 x 10^{14}	3 x 10^{12}
Pulse duration (μsec)	~ 1	~ 0.3
Beam energy per pulse (kJ)	100	12
Energy deposited in Hg per pulse (kJ)	~ 60	6
Expected peak energy density in Hg (MJ/m^3)	47	3
Peak local temperature rise in Hg (K)	26	1.6
Peak pressure increase in Hg (MPa)	105	7.1

Such a sudden power deposition in the incident window of the target container and the target material produces pressure waves which attack the target container, especially the incident window. As a simple case, if we assume a cylindrical Hg target of 20 cm in diameter with a semi-spherical dome window as shown in Fig. 5, calculated values of maximum stress levels at the center of the incident window and at a typical point on the cylindrical surface reach at the values listed in Table 3 [15] and the time behaviors at the window center become as shown in Fig. 6 [15]. A rectangular (uniform) beam density distribution gives the highest stress level than other distributions.

In order to validate the calculations, measurements of pressure waves in an Hg target and resulting stress in an target container are being in progress under an international collaboration using the AGS proton synchrotron at Brookhaven National Laboratory. Figure 7 shows measured deformation velocity at a point of the cylindrical surface of the container [16], due to the stress caused by pressure wave. The data is still very preliminary, but the JAERI team was successful to detect such data using a Doppler Laser instrument developed at JAERI and it was confirmed that the calculated result was very close to the measurement, although there exist small discrepancies between them Since in the container used in the experiment, there were many small flanges, etc. around the cylindrical surface, the measured condition was somewhat different from the ideal calculational model. Nevertheless, it can be said that the stress level and timing of the first peak is well reproduced by the calculation. Thus, the use of the present calculation codes, model, assumptions, etc. seems to be acceptable, at least in the first order estimation.

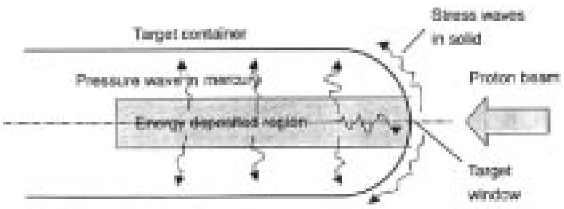

Fig. 5 Concept of pressure wave propagation in target container

Table 3 Approximate level of stress in target container window and vessel for different beam-density distribustions

Beam profile	Max stress on window (MPa)	Max stress on cylinder (z=12 cm)*
Uniform	170 (110)	136
Gaussian	100 (80)	110
Parabolic	100 (90)	120
Moffett	0 (90)	120 - 140

*12 cm from incident surface

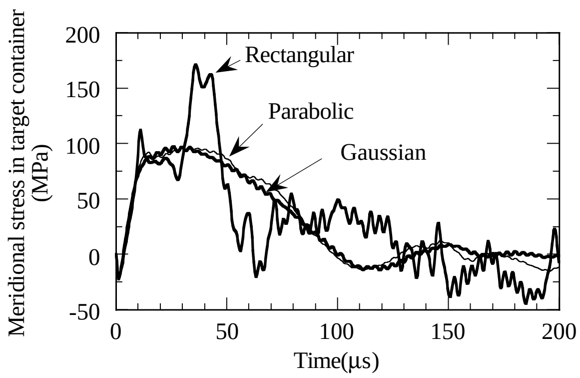

Fig. 6 Meridional stress changes at the center of window under various beam profile; rectangular, paraboric and Gaussian

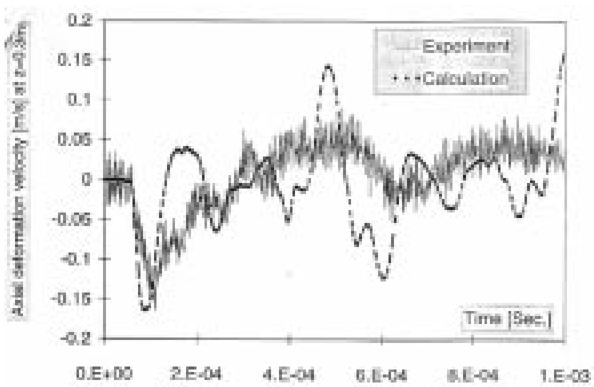

Fig. 7 Measured and calculated deformation velocity

The calculated values of the stress levels in the target container are to be compared to those of the allowable design stress of a typical candidate material for the target container: For instance, for SUS316, 137-110 MPa (at 423-673 K) for membrane + bending and 200-160 MPa (at 423-673 K) for membrane. In case of an uniform beam distribution the maximum stress level exceeds the allowable design stress, while in case of round distributions as Parabolic one, the level is below the allowable design stress. However, in more realistic case, a flat target, higher stress levels will be brought about. It reaches even at about 480 MPa, which is much higher than the allowable design stress, at a flat surface, if we assume a cavitation effect of the pressure wave. Therefore, an effective way to mitigate such pressure wave must be established. Skala, et al. have proposed to inject small helium gas babbles into Hg [17]. In addition to the dynamic stress mentioned above, there is a static stress, due to a static temperature difference in the window, which is 90-100 MPa under the beam current density of 48 $\mu A/cm^2$.

The effect of radiation damage on the mechanical properties of candidate materials must be considered under such stress and coolant (target material) flow conditions at the service temperature. The frequency of the dynamic stress is at least 50 Hz, the repetition rate of the pulsed proton beam, but if we take into account successive reflected waves as shown in Fig. 7, it becomes to an order of 20 kHz, Characteristics of materials for such *very high frequency fatigue* have not been well known.

6. Radiation damage

In an intense pulsed spallation source a proton beam window is subject to the most serious radiation damage, in which we must consider the effects not only of fast neutrons below 20 MeV, but also of those above 20 MeV and incident protons. For the projected JAERI source (1.5 GeV protons, 3.33 mA (5 MW), 48 $\mu A/cm^2$ in current density), dpa (displacement per atom) values for a typical candidate material (stainless steal) have been calculated [18], which are summarized in Table 4 compared with corresponding values estimated for SNS [19] and [20] ESS. Values estimated for the first wall of a fusion reactor (CTR with 1 MW/m^2) are also shown for reference.

Table 4 Comparison of important radiatopm damage parameters in the incident proton beam window in intense spallation sources

	dpa/y				Helium appm He/y	Hydrogen appm H/y
	Neutrons (E < 20 MeV)	Neutrons (E > 20 MeV)	Protons	total		
JAERI (5 MW)	39.45	6.89	21.16	67.5	2,270	
SNS (1 MW)	~ 12.5	~ 1.4	~ 8.9	22.8	1,014	10,840
ESS (5 MW)				~ 60	4,500	70,000
CRT (1 MW/m^2)				18	250	800

Although there are some differences in the dpa values per MW (dpa/MW) between the three spallation sources, due to different proton energies, accordingly different proton currents, different calculation codes and different assumed maximum current densities, those values are more than three times as large as a corresponding value in the first wall of a fusion reactor. As far as radiation damage in terms of dpa is concerned, the contribution from protons is only about one third of the total in the spallation sources. However, the material property change is very much different from those in a fusion reactor and a fast reactor, even for the same dpa. For example, the production rates of helium (He) and hydrogen (H) per dpa in the beam window of a spallation source are one order of magnitude and about 30 times larger than those in the first wall of a CTR, respectively. Note that the corresponding values for a fast fission reactor are negligibly small compared to the above mentioned values. Such large He and H production rates cause serious material hardening and He/H embrittlement. It is still not clear whether all hydrogenous produced can be retained in the material. Some experiments by triple beam bombardment indicated that a large amount of H diffused out from the surface at elevated temperatures. It must be confirmed by experiments whether such a H diffusion can be expected, even in a much deeper H distribution in an actual beam window, . In the ESS project a higher operating temperature of a Hg target is tentatively assumed expecting such effects.

In order to accumulate data base for radiation damage of some important materials by proton beam irradiation, extensive irradiation experiments are under progress by using 800 MeV protons from LAMPF proton linac and 560 MeV protons at PSI (installed in the Zircaloy target of SINQ) and some preliminary data are being available. Some irradiated samples are available from used (spent) components; for example, Inconel 718 from a beam window used at LAMPF [21], Ta from ISIS target, etc. Most of those samples were irradiated at natural temperature, and so on. Experiments under more realistic environments (in a service temperature, under stress, in flowing Hg, etc.) are most important and such experiments using 72 MeV protons from the Injector-I cyclotron at PSI and 600 MeV protons from the proton linac at Moscow Meson Factory are under planning. JAERI is partly participating in the present irradiation program at PSI but more active participation is highly required in very near future.

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