



# Summary of third workshop on materials science and technology for the spallation neutron source at KEK, March 2002

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## Abstract

The third workshop on the Materials Science and Technology for Spallation Neutron Source was held on 14–15 March, 2002 at KEK to discuss the present status and problems concerning the material issues for the spallation neutron source designs and associated R&D. There were 62 in attendance. The following six topics were energetically discussed: general presentations, radiation damage, solid target, cladding techniques, corrosion tests and accelerator-driven systems. Conclusions were reached in several areas: the basic technology of solid targets, including a Ta-clad tungsten block with the HIP method might be well-established, but irradiation and fatigue data are not yet sufficient. Liquid-target technology seems to have several problems: the urgent problem of mercury targets for neutron-scattering facilities is pitting by pressure waves induced by pulsed protons. The PIE of the STIP experiments is expected to prove the irradiation data. Problems of materials and coolant under irradiation, such as IASCC, should be investigated with the STIP and ion irradiation. Further, R&D for a lead–bismuth target is needed to design an ADS.

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## 1. Introduction

*The Workshop on Materials Science and Technology for Spallation Neutron Source* has been held at KEK every fiscal year by both the Target Developing Team (of KEK and universities) and the Target Subgroup in the Neutron Group of the JAERI–KEK Joint Project for High Intensity Proton Accelerators (acronym of the project: J-PARC) since the spring of 1999 when MOU (Memorandum of Understanding) for the Joint Project was exchanged between KEK and JAERI. The first workshop was held on 9–10 December, 1999, while treating six kinds of sessions: plenary (3), designs of spallation targets and associated material issues (4), mercury target R&D (5), solid target R&D (4), radiation damages including computer simulation (4) and material irradiation experiments (3). The number of presenta-

tions is shown in each parenthesis. In all, 23 oral presentations were made and discussed among 42 participants [1]. Discussions were concentrated on R&D issues and research plans on spallation targets and materials. There were also several presentations on experiences in fusion materials. The second workshop was held on 21–22 March, 2001. Invited participants were S.A. Maloy of LANL, P. Ferguson of SNS and H. Takahashi of BNL [2]. There were a total of 84 participants and 8 sessions: General (3), high-energy nuclear reactions (3), liquid target R&D (4), solid target R&D (7), corrosion test by water and Pb–Bi (3) and radiation damage (7). In each session, the results of R&D work were presented from each team.

The third workshop was held on 14–15 March, 2002. There were 62 people in attendance, including Yong Dai from PSI [3]. The following six kinds of topics were discussed: general presentations (3), radiation damage (6), solid target (2), cladding techniques (3), corrosion and fatigue tests (3) and accelerator driven systems (4). A summary of the presentations and discussions follows.

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## 2. Contents

The program of the workshop is given in Table 1. Preceding the general session, M. Kawai made an

opening remark about successful HIP'ing of a tantalum-clad target block for KENS as well as a brief presentation on current status of the R&D carried out by the Target Developing Team.

Table 1

Program of Third Workshop on Materials Science and Technology for Spallation Neutron Sources

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Date: 13:10–18:40 14 March, 9:00–17:00 15 March, 2002

Place: Seminar Room Nos. 1 and 2, 2nd Floor, Building 4, KEK

-Presentation-

14 March, 2002

13:00–13:10 Opening

0.1 Greeting (S. Ikeda, KEK)

0.2 Opening Remarks (M. Kawai, KEK)

13.20–15.00 General Session <N. Yoshida, Kyushu Univ.>

1.1 Status Review of the Spallation Neutron Source Development (M. Furusaka, KEK)

1.2 Performance of Cold Neutron Source and Its Material Issues (N. Watanabe, JAERI)

1.3 Report on Third International Workshop on Spallation Neutron Source (K. Kikuchi, JAERI)

<Coffee Break>

15:15–17:15 Radiation Damage (1) <A. Hishinuma, JAERI>

2.1 Material irradiations and PIE at SINQ (Y. Dai, PSI) (1 h)

2.2 Ion Irradiation for A Simulation Experiment of Target Materials for SNS (E. Wakai, JAERI)

2.3 Irradiation Behaviors of Tungsten by HVEM (T. Yoshiie, KUR)

17:15–18:45 Radiation Damage (2) <S. Uchida, Tohoku Univ.>

2.4 Irradiation Hardening of Tungsten and its Alloys after Helium- and Hydrogen-ion Implantation (A. Hasegawa, Tohoku Univ.)

2.5 Irradiation Assisted Stress Corrosion Cracking of type 316 stainless steel in High Purity Water (Y. Miwa, JAERI)

2.6 Post-irradiation examination of materials irradiated by high energy neutron (N. Yokota, Nippon Nuclear Fuel Develop. Co., Ltd.)

<Dinner Party> at Onomura

15 March, 2001

9:00–10:00 Solid Target <R. Hino, JAERI>

3.1 Thermo-hydraulic Designs of MW-class Solid Target (K. Mishima, KUR)

3.2 Neutron Performance of Solid Target (D. Nio, Hokkaido Univ.)

<Coffee Break>

10:15–11:45 Cladding Technique <T. Igarashi, Osaka Prefecture Univ.>

4.1 Frontiers of Material Coating Technique (N. Kitagawa, Sumitomo Metal Mining Co. Ltd.)

4.2 Joining of Ta-Sheet to W-Block by a Brazing Technique (Y. Hiraoka, Okayama Univ. of Science)

4.3 Compact Coating of Tantalum on Nickel by Molten Salt Electro-Deposition (M. Mehmood and T. Yamamura, Tohoku Univ.)

<Lunch>

13:00–14:30 Corrosion and Fatigue <S. Jitsukawa, JAERI>

5.1 Corrosion Resistance of Tungsten and Its Alloys for Spallation Target in Aqueous Solution (K. Sugimoto, Tohoku Univ.)

5.2 Corrosion Resistance of Refractory Metals in High Temperature Water (H. Kurishita, Tohoku University)

5.3 Fatigue Data of 20kHz Supersonic Fatigue Test (N. Komine, Shimadzu Corporation)

<Coffee Break>

14:45–16:45 ADS Target <Y. Kiyanagi, Hokkaido Univ.>

6.1 Accelerator Driven System for Nuclear Transmutation and Its Target (H. Takano, JAERI)

6.2 Present Status of R&D on Lead–Bismuth Loop Experiment at JAERI (S. Saitoh, JAERI)

6.3 Experiment Study on Lead–Bismuth Flow Technology and Steel Corrosion (M. Takahashi, Tokyo Institute of Technology)

6.4 200 kW Pb–Bi Target and Irradiation Test by Using It (M. Futakawa, JAERI)

16:45–17:00 Closing Session

7.1 Summary Talk (Hiroaki Kurishita, Tohoku Univ.)

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### 2.1. General session

M. Furusaka of KEK introduced the importance of the new neutron scattering facility 'JSNS' in the field of material and life science for the JAERI-KEK Joint Project of High Intensity Proton Accelerators. He then described its current status. Fig. 1 shows the accelerator complex of the project. For JSNS, 23 neutron instruments have been tentatively selected from the originally proposed 40 instruments in order to design a target-moderator-reflector assembly (TMRA). The target system of the JSNS will be a cross-flow type mercury target. Studies on the instrument site and their requirements resulted in the decision to install three supercritical hydrogen moderators at 20 K (one decoupled and poisoned, and one coupled), all with two-surface neutron-extraction, as shown in Fig. 2. Neutronic optimization of the moderator gives a coupled moderator whose neutron intensity will be about three-times higher than that of SNS at the pulsed peak. Nearly 50% of the instruments will be located at the coupled moderator beamline. Pulse-shape optimization designs were made for the decoupled and poisoned moderators as well as the reflector, of which the material was changed from lead to a complex of beryllium and iron. R&D work is on-going concerning the mercury target system; a flow simulation analysis compared with a full-scale water flow mock-up test, a corrosion/erosion test, and an impact erosion test to solve pitting problems due to short-pulsed proton beam deposits with very high power. A remote-handling test bench has been built and operated to study target exchange together with computer simulations. Various device-developing studies are also going on for neutron detectors and instruments.

N. Watanabe of JAERI presented the material issues of a cryogenic moderator to provide a long life and an intense and sharp pulsed neutron source. A new de-

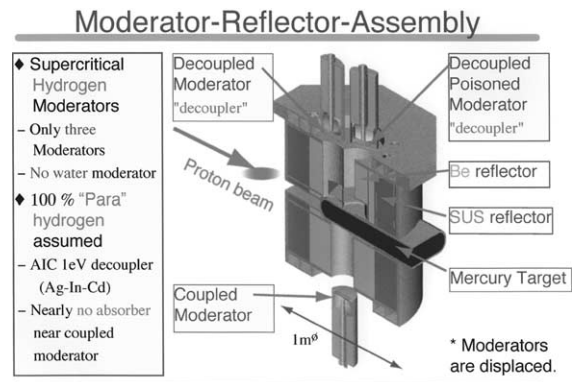


Fig. 2. Target moderator-reflector-assembly of JSNS. In the figure, all moderators are extracted from the normal positions which are very close to the mercury target during operation.

coupler of a Ag-In-Cd complex was designed. Fig. 3 shows that the neutron transmission of the new decoupler resembles that of the familiar B<sub>4</sub>C. However, its fabrication method has not been established and its R&D work has been discussed.

K. Kikuchi reported on the 3rd Workshop on Mercury Target Development held at ORNL last November. There were sessions concerning mercury target issues and the target system design. The main concerns were pitting on the target vessel container with relevance to cavitation damage caused by pressure waves. Pitting was firstly found in an off-beam test with a Split Hopkinson Bar testing machine at JAERI in 2000. It was secondary found in an in-beam-test after only 100 pulses in WNR at LANL. For the real target, the number of pulses at 25 Hz repeat is over  $2 \times 10^8$  during one half of a year. Some pitting will be likely to have a depth a couple of tens of mm, whereas the vessel thickness is designed to be 2 mm. At the workshop, pitting phenomenon, new testing to clarify the influence on a real target and mitigating

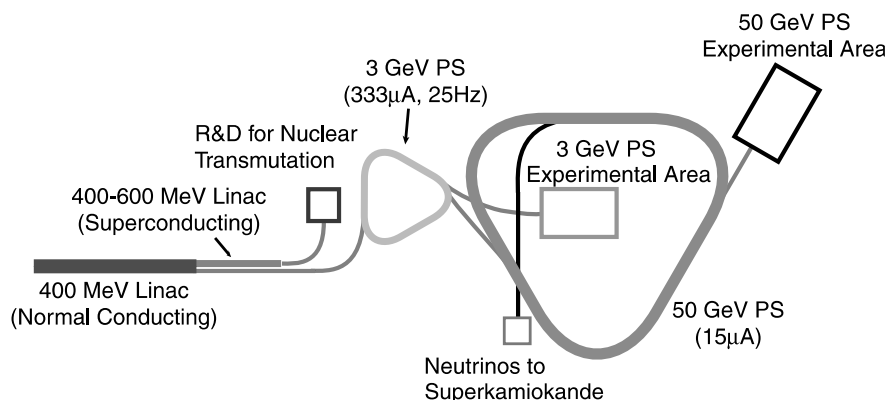


Fig. 1. Accelerator complex of the JAERI-KEK Joint Project of High Intensity Proton Accelerators (JPARC project). Neutron scattering facility is in the 3 GeV experimental area.

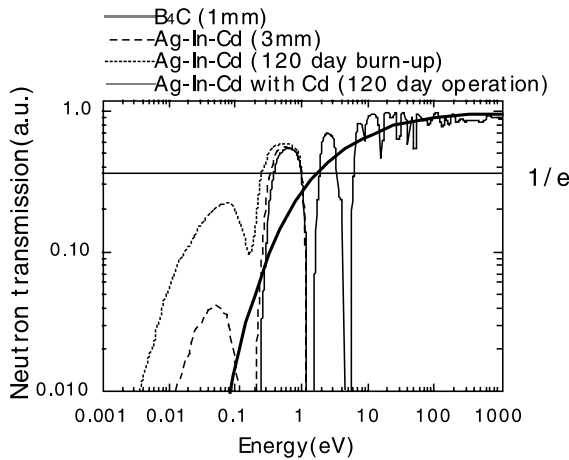


Fig. 3. Comparison of the neutron transmission of various decouplers.

methods were discussed, and near-future work was proposed.

## 2.2. Radiation damage

Data of radiation damage is urgently required for target designs of several new projects (SNS, JSNS and ESS). The SINQ target irradiation program, the STIP experiment and PIE, was introduced by Y. Dai of PSI. From PIE of the STIP-I experiment, interesting data

were obtained for the window and target vessel materials. Fig. 4 shows the PIE results for F82H irradiated in the STIP-I Experiments. For the STIP-II and -III experiments, more information will be obtained for the solid target and the liquid-target-and-vessel complex with higher irradiation.

E. Wakai presented irradiation experiments with a triple beam of H, He and Fe on F82H material. The swelling of ferritic/martensitic steels under triple beams was larger than that under dual beams from 500 to 600 °C, and hydrogen strongly influenced the swelling behavior. However, it was discussed why hydrogen could not be detected from a sample implanted to 4–8 at.% by hydrogen, even by means of  $^1\text{H}(^{15}\text{N}, \alpha\gamma)^{12}\text{C}$ , and to where hydrogen goes.

T. Yoshiie and A. Hasegawa presented the results of irradiation experiments with electrons and light ions ( $\text{H}^+$ ,  $\text{He}^+$ ) on tungsten and its alloys. It is interesting that in different kinds of samples, the order of the number of interstitial dislocation loops produced by electrons was the same as that of irradiation hardening induced by ions. Both results of microscopic observations with TEM and macroscopic property of hardening showed us which tungsten has strong irradiation resistance as well as fundamental information on the irradiation mechanism. In these experiments, the order of the resistance was  $\text{PFW} < \text{CVD-W} < \text{5N-W}$  (99.999% purity).

Y. Miwa's presentation described to us the need of irradiation experiments to clarify the irradiation assisted

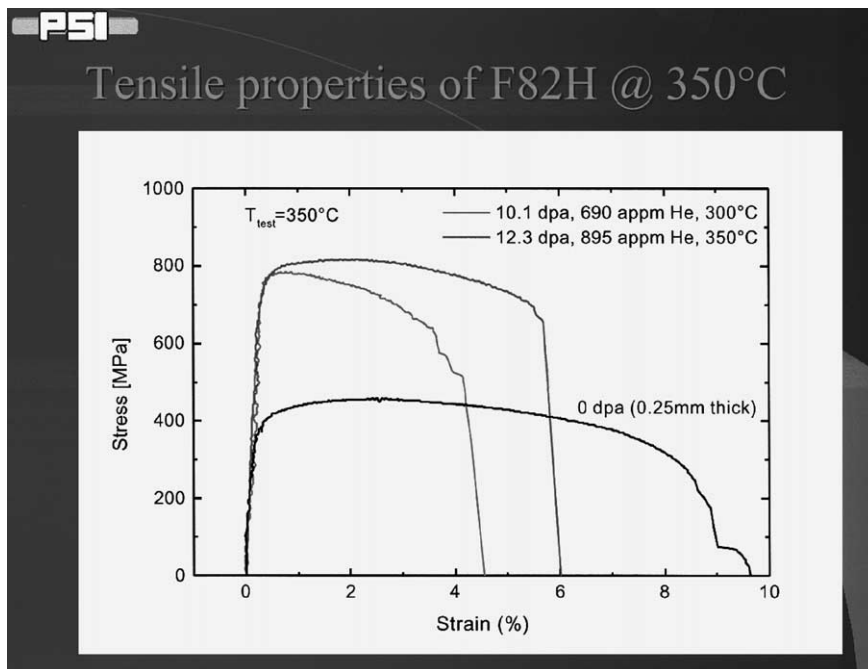


Fig. 4. Tensile properties of F82H measured in STIP-I experiment.

stress corrosion cracking (IASCC) problem in spallation materials in contact with coolant water.

N. Yokota of NFD introduced the hot laboratory of his company and described a post-irradiation examination for nuclear fuels in a boiling water reactor and reactor materials in fission reactors, and the R&D of reactor structural materials for plant-life extension and preventive maintenance. Fig. 5 shows a typical example of TEM observations of dislocation loops, showing the recovery of damage for type 304SS. Dislocation loops decrease with an increase of the annealing temperature. The laboratory can treat very high activity samples. Accordingly, the PIE of high activity specimens from the STIP-experiments would be carried out in this laboratory.

### 2.3. Solid target

K. Mishima and D. Nio presented thermal-hydraulic designs of two types of solid targets composed of tungsten up to 5 MW and their thermal-and-cold neutron performance, respectively. Table 2 gives the ratios of the neutron intensities for the rod-type tungsten target with a tantalum sheath at 1 MW proton beam power to the mercury target. The optimized solid target is predicted to have a better neutron performance than the mercury target up to at least 1 MeV. Nio also showed that the back-up solid target, which was substituted for the mercury one of JSNS without any optimization of moderator configuration, had almost the same neutron performance as the mercury target. From the view point

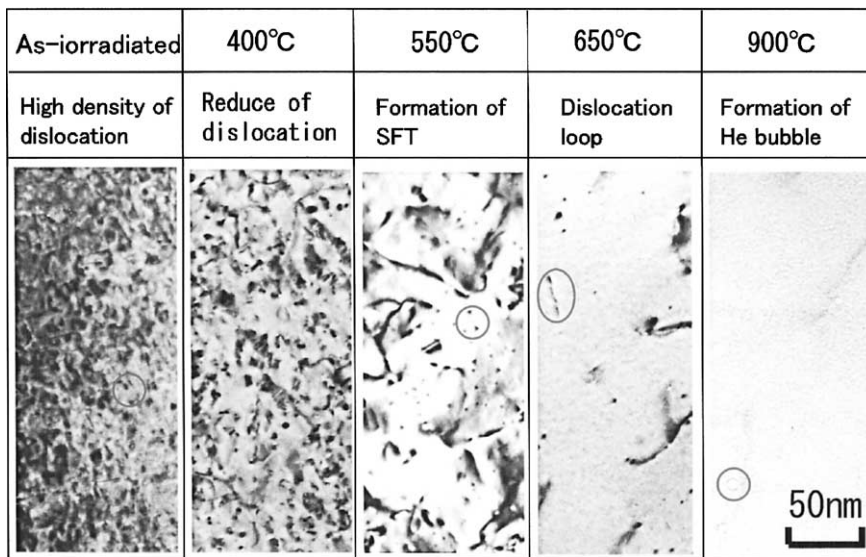


Fig. 5. TEM observation of the recovery of damage for type 304 SS. Irradiated by  $1.4 \times 10^{26}$  n/m<sup>2</sup> and heated for 1 h.

Table 2

Ratio of the neutron intensities in the case of a rod-type tungsten target with a tantalum sheath

Moderator	Energy interval (meV)	Ratio of integrated neutron intensity to that of mercury target		
		Dw = 1.0 mm (1.07)	Dw = 0.5 mm (1.17)	Dw = 0.0 mm (1.26)
Coupled moderator	0–5	0.97	1.03	1.09
	5–25	0.97	1.03	1.08
	25–100	0.98	1.04	1.09
	100–500	0.98	1.06	1.10
Poisoned moderator	0–5	0.93	1.02	1.06
	5–25	0.94	1.03	1.08
	25–100	0.95	1.04	1.08
	100–500	0.91	1.02	1.03

N.B. Dw is a diameter of wrapping wire around a rod to keep a separation of rods. The wrapping wire will cause a circulation flow of water around the rods and promote cooling of the rods.

Values in parenthesis denote material density ratio.

of engineering, the rod-type target would be preferable to a slab target of tantalum-clad tungsten because the fabrication techniques for rods have been established in the nuclear reactors. Of course, it is necessary to investigate the reliability of the solid target in long-term thermal cycling and irradiation.

2.4. Cladding technique

A CrN film presented by N. Kitagawa showed a high corrosion-and-wear resistance and excellent oxidation resistance due to the high hardness. In a corrosion test for valve seats, CrN shows the remarkable result that the pin-hole density decreases logarithmically with the CrN film thickness, as shown in Fig. 6. From experiments, it was concluded that a CrN coating could be applied to many parts that require corrosion and wear resistance in a nuclear reactor. It was also suggested to guard against pitting on the window of the mercury target mentioned above.

Y. Hiraoka and M. Mehmood presented the techniques of tantalum cladding on tungsten by means of brazing and an electrolytic molten-salt deposition, respectively. They succeed to conduct fundamental experiments on cladding. A further investigation will be needed to establish the technique as an actual application to a target. The HIP'ing method is considered to be established to fabricate the tungsten target block, as shown in the case of the KENS target. The electrolytic molten-salt method is a very new one and is expected to be used to fabricate the target economically in the future.

2.5. Corrosion and fatigue tests

K. Sugimoto and H. Kurishita presented experiments on the corrosion resistance of tungsten and heavy metal.

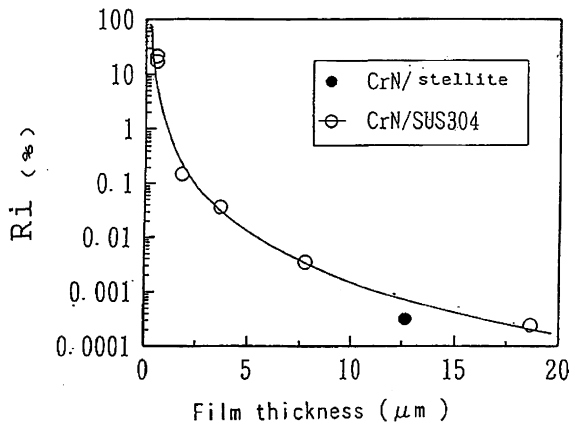


Fig. 6. Relation between the CrN film thickness and the pin-hole density (Ri).

The former experiments were made in an aqueous solution at 100 °C, while changing the condition of the flow rates and hydrogen contents in water. Fig. 7 shows that the mass loss of tungsten remarkably increases along with the flow rate. A higher purity tungsten, such as a CVD-W (99.9999% purity), showed a higher corrosion resistance at a low flow rate, but it experienced a larger mass loss at a high flow rate, because of erosion by the flow. The latter shows that the weight loss of PFW (99.95% purity) at 180 and 260 °C is as low as 0.022 mm/y. However, Sugimoto's experiments indicated that cladding will be necessary for the tungsten of a MW-class target needing high-velocity cooling. It was surprising that CrN-coated tungsten still showed excellent resistance, even at 320 °C, where the other materials were heavily corroded.

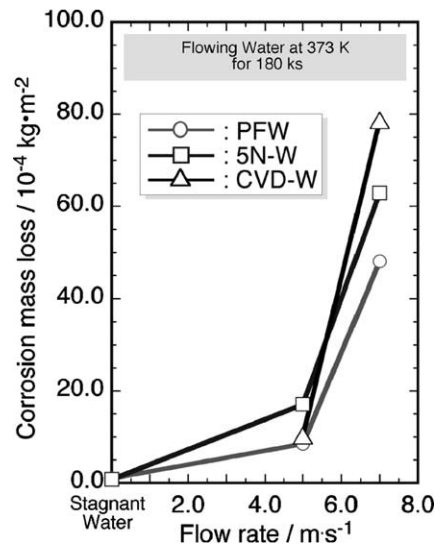


Fig. 7. Corrosion mass loss of W and W alloys in stagnant and flowing water saturated by H<sub>2</sub> at 100 °C for 180 ks.

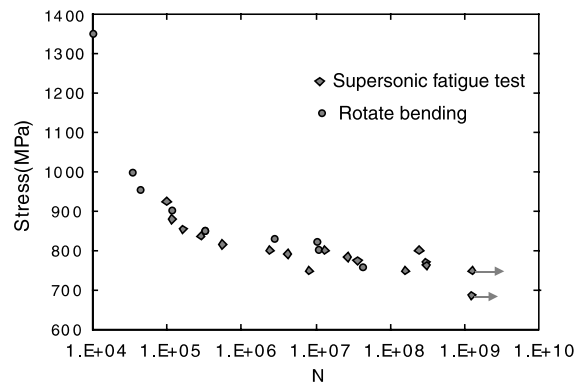


Fig. 8. Fatigue test results for SNCM439 steel.

Fatigue tests are needed for both unirradiated and irradiated spallation materials. The resonant dumbbell specimens presented by N. Komine of Shimadzu Corporation will enable us to conduct a fatigue test at a

frequency of 20 kHz. In his experiments, the fatigue limits with dumbbell specimens were in agreement within about 10% with the data of the rotary bend testing, as shown in Fig. 8.

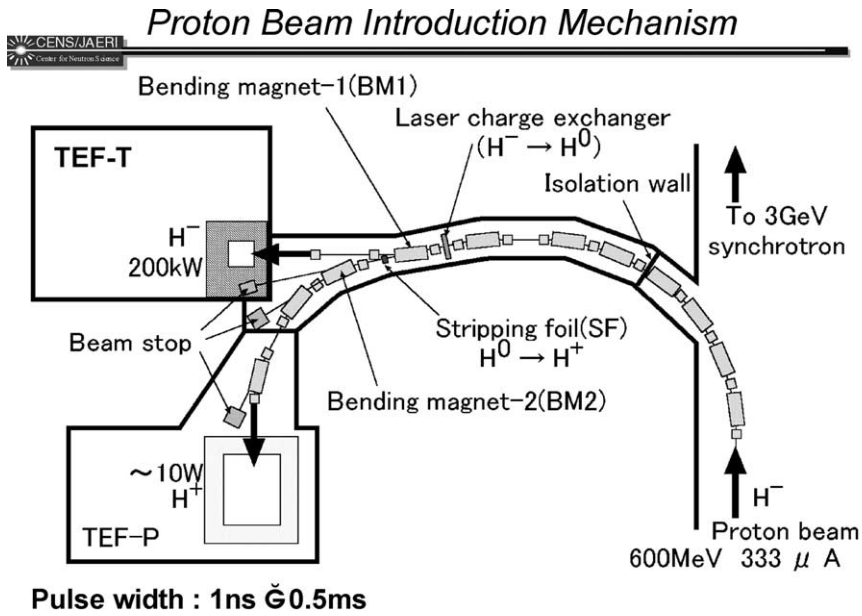


Fig. 9. Transmutation experimental facility of joint project.

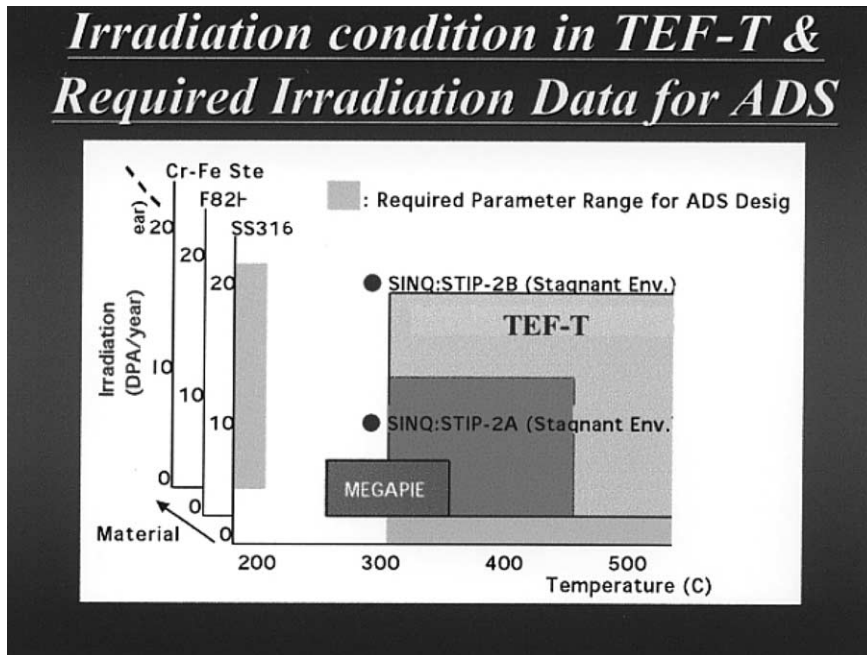


Fig. 10. Comparison of the irradiation condition of TEF-T and MEGAPIE.

### 2.6. Accelerator driven system

H. Takano gave a brief description of an accelerator-driven system (ADS) for nuclear transmutation and JAERI's activities on conceptual designs of the ADS systems, and the experimental program for the development and demonstration of accelerator-driven transmutation technology under the project plan of the High Intensity Proton Accelerator. Fig. 9 shows the transmutation experimental facility (TEF) for the development of the ADS technology to be constructed in the Joint Project. M. Futakawa, showed the scope of the ADS Target Test Facility with a 200 kW Pb–Bi Target bombarded by a pulsed proton beam at 600 MeV, 25 Hz. He also explained the irradiation conditions compared with those of the STIP experiments and MEGA-PIE Project, as shown in Fig. 10.

S. Saito and M. Takahashi independently presented experiments carried out in lead–bismuth loops. Saito's experiment was made at a maximum temperature of 450 °C, a Pb–Bi velocity at the test section of 1 m/s and the estimated oxygen content,  $1.5 \times 10^{-7}$  wt%. The test section was made of a type 316 stainless steel tube. The results showed a specimen surface containing several pits and a decrease of the tube thickness by 0.1 mm/3000 h. From Takahashi's experiment, the following results were obtained: after exposure for 1000 h of steel to a Pb–Bi flow at 550 °C, 2 m/s and an oxygen concentration of  $3.6 \times 10^{-7}$  wt%, serious erosion damage was observed in SCM420 at the entrance; some erosion damage appeared in low-Cr steels (SCM420, F82H, STBA26 and HCM12) downstream. Penetration of Pb and Bi into materials was also observed in SCM420, STBA26, STBA28 and SS316.

### 3. Conclusion

In the workshop, many results from R&D were energetically discussed. Conclusions were reached in several areas: the basic technology of solid targets including a Ta-clad tungsten block with the HIP method might be well established, but the irradiation and fatigue data are not yet sufficient. Liquid target technology seems to have several problems: the urgent problem of mercury targets for neutron-scattering facilities is pitting by pressure waves induced by pulsed protons. It should also be investigated whether a similar problem will occur in a solid target. PIE of the STIP experiments is expected to prove the irradiation data. Problems of the materials and the coolant under irradiation, such as IASCC, should be investigated with the STIP and ion irradiation. The techniques of ceramic coating and fatigue testing will be also useful for spallation material technology. Further R&D for a lead–bismuth target is needed to design the ADS.

### References

- [1] M. Kawai, K. Kikuchi (Eds.), Workshop on the Materials for Spallation Neutron Source, KEK Proceedings 2000-18, August 2000 (in Japanese).
- [2] M. Kawai, K. Kikuchi (Eds.), Proceedings of Second Workshop on the Materials Technology for Spallation Neutron Source, KEK Proceedings 2001-11, June 2001.
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