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Materials irradiation facilities at the high-power Swiss proton accelerator complex

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Abstract

Within the Swiss proton accelerator complex at the Paul-Scherrer-Institute (PSI), several irradiation facilities are operated for investigation of materials behavior under high-dose irradiation conditions as well as for neutron activation analysis and isotope production. In LiSoR (liquid solid reaction), a liquid metal loop connected to the 72 MeV proton accelerator Injector 1, steel samples are irradiated while being in contact with flowing lead–bismuth-eutectic (LBE) at elevated temperatures and under tensile stress. In the spallation neutron source SINQ, the STIP program (SINQ Target Irradiation Program) allows materials irradiation under realistic spallation conditions, i.e. in a mixed spectrum of 570 MeV protons and spallation neutrons. Hundreds of samples, mainly austenitic and ferritic–martensitic steels such as 316L, T91 or F82H, were irradiated to doses up to 20 dpa as part of STIP. These also included steel samples in contact with liquid Hg and liquid LBE. MEGAPIE (MEGAwatt PIlot Experiment), a liquid metal target employing LBE, operated in SINQ during the second half of 2006, can be taken as a materials irradiation facility on its own. Adjacent to the target position, SINQ houses a neutron irradiation rabbit system serving activation analysis and isotope production.

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1. Introduction

Despite decades of experience in reactor technology and accelerator developments, materials in high-dose irradiation environments are still an issue of widespread interest. Related to that, there is worldwide an increasing demand for appropriate materials irradiation facilities. The demand stems from different disciplines: Target development initiatives for spallation neutron sources or neutrino factories, new fission reactor lines (Generation IV) and also fusion reactor technology. Their common requirements are to accumulate a meaningful dose, i.e. a sufficient number of dpa's (displacement per atom) during a tolerable irradiation period, from a correct spectrum of irradiating particles and at the correct temperatures of interest. The latter two requirements are partly diverging from case to case: whereas spallation sources have to deal typically with a mixed spectrum of protons and neutrons, the new fission reactor lines require a spectrum of mainly fast neutrons with no protons, and the fusion technology is heading for a 14 MeV neutron spectrum. The temperature requirements range

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from just above room temperature (spallation sources) to high temperatures up to and above 1000 °C (Generation IV).

In addition to a relatively limited number of operating and accessible irradiation facilities, three dedicated large irradiation facilities are presently under consideration or in the project planning phase: The Los Alamos Material Test Station (MTS) that projects a 1 mA/800 MeV proton beam spallation target in a sandwich structure with the irradiation specimens placed in the centre in a spectrum of primarily fast neutrons (goal: 10^{15} n/cm^2/s); IFMIF (International Fusion Materials Irradiation Facility) that is dedicated for fusion reactor materials irradiation [1]; the Jules-Horowitz (JH) reactor project [2]. For all three, the final decisions for funding and the dates of realization are still open. MTS has a chance to start in 2008. For IFMIF and JH the estimated budget requirements are between 500 and 1000 M€, and, if funded, the earliest completion date is after 2015. On a short or intermediate time frame this scenario is not at all very encouraging.

PSI operates a proton accelerator complex, combining a 72 MeV injector and 590 MeV ring cyclotron with a final beam power in the MW range: a 1.8–2 mA proton beam is routine at present, and upgrades to ultimately 3 mA are planned. The first two targets in the proton beam line are graphite targets for meson (muon and pion) production, followed by the final heavy metal target used for the spallation neutron source SINQ [3]. The time structure (51 MHz) of the proton beam is lost after hitting the spallation target, thus, SINQ is effectively a continuous spallation source at the MW beam power level.

The PSI accelerator complex, including SINQ, offers various options for materials irradiation to high doses in proton and/or neutron beam environments. The stations currently in use are the followings: (i) The LiSoR (liquid solid reaction) facility at the (old) Injector 1 beamline position, which receives a 72 MeV proton (or on demand He or heavier ions) beam of small (mm) size and max 50 μA/cm^2 of beam current density, (ii) STIP, the SINQ Target Irradiation Program, where the SINQ target sample rods in the central reaction zone are used to irradiate material specimens by a mixed spectrum of 570 MeV protons and spallation neutrons, (iii) a neutron irradiation rabbit system, transporting sample capsules to positions inside the D_2O moderator vessel in close vicinity of the spallation target. MEGAPIE (MEGAwatt Pilot Experiment), a liquid metal target employing lead–bismuth-eutectic (LBE), operated in the second half of 2006, may be considered as an irradiation facility on its own. In particular, the beam window deserves paramount attention: Fabricated of ferritic–martensitic steel T91 (9Cr2WTaV), it received the full load of a MW scale proton beam in contact with an inner circulating liquid LBE.

The present paper aims to compile specific details on these irradiation facilities, and elucidate their potentials (and limitations) in view of materials irradiation needs of the interested communities.

2. LiSoR

As mentioned in the introduction MEGAPIE used LBE as the spallation material and the ferritic–martensitic steel T91 as the beam entrance window. LiSoR was launched as a supporting R&D facility for MEGAPIE to investigate the simultaneous influence of flowing LBE at elevated temperatures, mechanical stress and irradiating protons on steel T91 to gain predictive information on the behavior of the steel under MEGAPIE relevant conditions.

LiSoR is an experimental LBE loop equipped with an electromagnetic pump, a flow meter and a heat exchanger system. Details of the LiSoR loop can be found in previous reports [4,5]. It has an exchangeable test section housing the test specimen, the whole loop being designed and constructed for operating in the environment of a proton beam bunker. Fig. 1 shows a photograph of a LiSoR test specimen, the dimensions in the proton beam area being 20 mm in width and 1 mm in thickness. A sketch of the test section is given in Fig. 2 showing the test tube (which houses the test specimen), the LBE in- and outlet, and the grip for the stress actuator. A real test tube, after having been irradiated and dismantled, is shown in Fig. 3. The photo was taken in the hot cell; the beam footprint is clearly visible at the lower half of the central test tube. It illustrates that, in addition to the test specimen on the inside, the test tube itself becomes an irradiation sample, the inside being in contact with liquid LBE and the outside in vacuum. Both samples were always examined. In addition, unirradiated reference material was available which was not in the proton beam but otherwise has experienced the same load and environment.

Although the beam energy of 72 MeV is considerably lower than that of MEGAPIE (570 MeV),

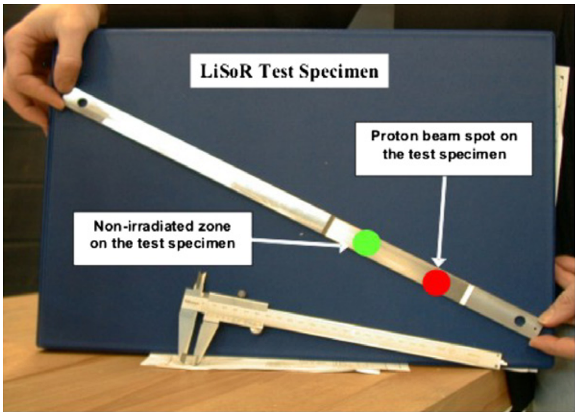

Fig. 1. The total length of the LiSoR test specimen is 495 mm. The area where the proton beam impinges the steel is marked by a spot.

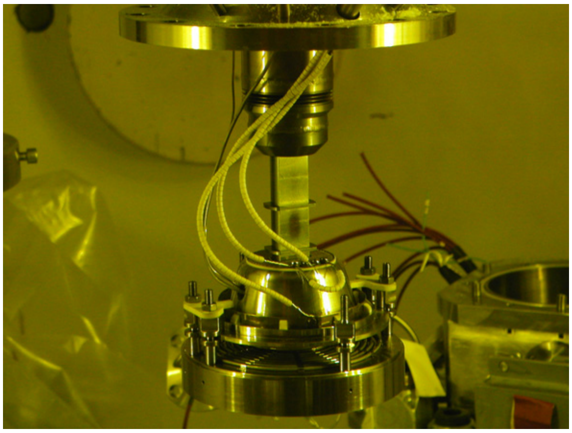

Fig. 3. LiSoR test tube after irradiation and dismantling (photo taken in the hot cell). The beam footprint is clearly visible at the lower half of the central test tube.

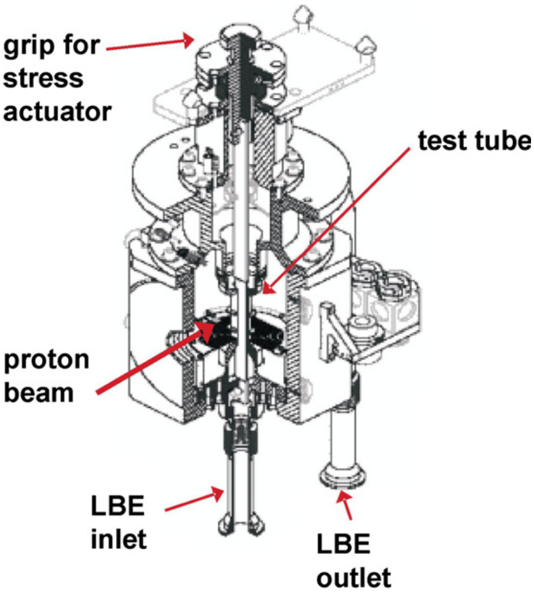

Fig. 2. Schematic showing a test section which houses the test specimen.

the flux at the sample position and hence the energy deposition in the material was adjusted to be of the same order of magnitude by focusing and wobbling of the beam.

Until now six irradiation cycles were performed. Table 1 gives an overview on the relevant parameters. Not all cycles ran as planned, in particular at the beginning. For example, a focused beam in combination with a too-low wobbling frequency caused a perforation of the test tube and leaking of LBE into the surrounding vacuum chamber. Another problem resulted from an imperfect leak detector design, signaling a non-existing leak which triggered the premature end to the experiment. Cycles 5 and 6 finally ran stable until the scheduled end. Detailed descriptions of each single test and the results of the irradiated material are given in [4–7].

One of the outcomes of the LiSoR experiments was the observation of an oxide layer of about 3 μm in thickness in the irradiated area formed during LiSoR cycle No. 2 [6,7]. In the oxide layer, two zones could be distinguished, a magnetite (outer layer) and a spinel (inner layer), illustrated in Fig. 4. The formation of oxide layers were also observed in the irradiated zones of samples exposed during LiSoR cycles No. 3, 4 and 5. It was found that the thickness of the oxide layers is dependent directly on the irradiation time and beam current (i.e. temperature). Adjacent to the irradiated area no oxide layer was observed.

This result is interesting (i) because in the reductive atmosphere of hydrogen (produced as a spallation product in LBE) oxide layer formation or even oxide layer stabilization is absolutely unexpected and (ii) the result is very promising in the sense of prevention of liquid metal embrittlement by the formation/stabilization of protecting oxide layers in the irradiated area at the beginning of the irradiation. We hope that this finding turns out to be sound in a more general sense, mitigating the problem of liquid metal embrittlement in irradiation environments.

In LiSoR run #6 coated specimens were irradiated but until now the test section has not been

Table 1
The beam parameters chosen for the six LiSoR irradiation cycles

LiSoR cycle	No. 1	No. 2	No. 3	No. 4	No. 5	No. 6
Beam energy	72 MeV	72 MeV	72 MeV	72 MeV	72 MeV	72 MeV
Proton beam	50 μA	50 μA	15 μA	30 μA	30 μA	30 μA
Beam profile on target	$\sigma_x = \sigma_y = 0.8$ mm	$\sigma_x = \sigma_y = 0.8$ mm	$\sigma_x = \sigma_y = 1.6$ mm	$\sigma_x = \sigma_y = 1.2$ mm	$\sigma_x = \sigma_y = 1.2$ mm	$\sigma_x = \sigma_y = 1.2$ mm
Beam wobbling	14.3 Hz in x 1.19 Hz in y (12:1)	14.3 Hz in x 1.19 Hz in y (12:1)	14.3 Hz in x 2.38 Hz in y (6:1)	14.3 Hz in x 2.38 Hz in y (6:1)	14.3 Hz in x 2.38 Hz in y (6:1)	14.3 Hz in x 2.38 Hz in y (6:1)
Duration of irradiation	1 h	34 h	264 h	144 h	724 h	882 h
Accumulated damage	~0.01 dpa	0.1 dpa	0.2 dpa	0.2 dpa	~1 dpa	~1.2 dpa

Irradiated area: wobbling horizontally $x_{max} = \pm 2.75$ m; wobbling vertically $y_{max} = \pm 7$ mm.

recovered from the loop because the activity is still too high. First results are expected to be available by the end of 2007.

All in all, despite of (or because of) unpredicted events valuable experience was gained by these experiments on strengths and weaknesses of systems combining the load of elevated temperature, stress and irradiation on a material in contact with circulating liquid metal, an experience indispensable for a safe operation and irradiation of MEGAPIE.

3. STIP program

For spallation targets in the MW-power range or accelerator driven systems (ADS), the irradiation induced degradation in the materials properties is a key parameter limiting the lifetime of targets. Because of the high displacement rate and production of spallation transmutation impurities, especially helium and hydrogen, the damage induced by spallation irradiation cannot simply be simulated by fission neutron irradiation and ion-beam implantation experiments. Such a demand triggered the development STIP [8,9], which offers the unique opportunity of high-dose irradiation under realistic spallation conditions. The program was conducted in collaboration with FZJ Jülich (D), LANL Los Alamos and ORNL Oak Ridge (USA), CEA (France), and JAEA (Japan), and has meanwhile irradiated several thousand samples of different materials of relevance (mainly steels) to doses up to 20 dpa.

In Target 3 (STIP-I), operated in the years 1998/1999, about 1500 miniaturized samples of different metals and steels and of different shapes for dedicated testing and examination purposes, were placed within the central target rods which receive the highest doses of protons and neutrons. The follow-up Targets 4 and 5 (STIP-II and -III) contained similar assemblies with irradiation samples. Fig. 5 shows the rod array of Target 4, along with a sketch illustrating the arrangement of the specimen rods. All targets were instrumented with thermocouples and dosimeter coupons to monitor temperature and dose at relevant irradiation positions. Based on this data, the individual dose and temperature of each sample could be computed. Fig. 6 shows flux distribution maps of protons and (thermal) neutrons as obtained from MCNPX calculations [10]. The relevant fluxes in the centre at the STIP sample positions for protons are in the 10^{14}/cm^2/s/mA

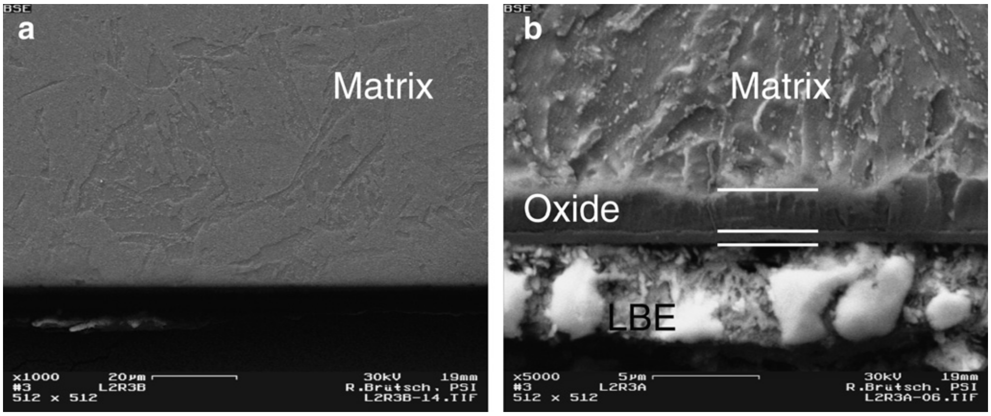

Fig. 4. Cross section of specimens recovered from LiSoR 2: (a) near, but outside the irradiated zone and (b) directly inside the beam foot print, the latter showing a protecting oxide layer on the steel surface (matrix), consisting of a magnetite layer on top and an intermediate layer of iron chromium spinel.

range, for fast neutrons between 3 and 7×10^{14}/cm^2/s/mA, and for thermal neutrons around 3×10^{13}/cm^2/s/mA.

Since its inception STIP has generated a huge amount of data [11–13] and received worldwide recognition from the spallation target, ADS and fusion technology community.

In particular, the results have been applied to the safety assessment of the MEGAPIE target [14]. As one example of relevance for MEGAPIE, Fig. 7 shows a photograph (taken in the hotlab) of a tensile sample and stressed capsule of martensitic steel 9Cr2WTaV irradiated in liquid LBE to 12 dpa in a temperature range of about 210–250 °C. Electron Probe Microanalysis indicates that the LBE corrosion and embrittlement effects are not severe under such irradiation conditions.

Without curtailing the success of STIP, the current sample configuration in the SINQ targets has two drawbacks:

- The irradiation temperature is mainly controlled by the beam heating, combined with the thermal contact to the surroundings. An independent temperature control of the irradiation samples would be highly desirable. In particular, higher temperatures than those achieved presently by beam heating are of increasing importance, in particular for the Generation IV developments.
- The period between sample definitions, preparation, loading, etc. until retrieving and analysing is very long, typically >4 years. If samples could be inserted into the target and could be taken out again in the operation position, without dismantling the target, the turnaround period could be significantly shortened.

A study has been launched for a modified STIP configuration aiming to overcome these drawbacks by a more flexible design for sample insertion and retrieval, combined with a controlled heating of the samples at the irradiation positions from the outside.

4. MEGAPIE

MEGAPIE [14] is a joint initiative by six European research institutions, the EU, JAEA (Japan), DOE (USA), and KAERI (Korea) to design, build, operate and explore a liquid lead–bismuth spallation target for 1 MW of beam power. The goal of this experiment is to explore the conditions under which such a target system can be licensed, to accrue relevant materials data beyond STIP and related programs for a design data base for liquid LBE targets, and to gain experience in operating such a system under realistic beam conditions. From the materials point of view, the beam window is of particular interest. The material chosen for the lower liquid metal container, which includes the beam window, is the martensitic steel T91. Fig. 8 illustrates the lower part of the target around the proton–LBE interaction zone, showing the lower liquid metal container and the surrounding double-walled safety hull, the latter made of the aluminum alloy AlMg3. A photograph of the real beam window is shown as well. The thickness of

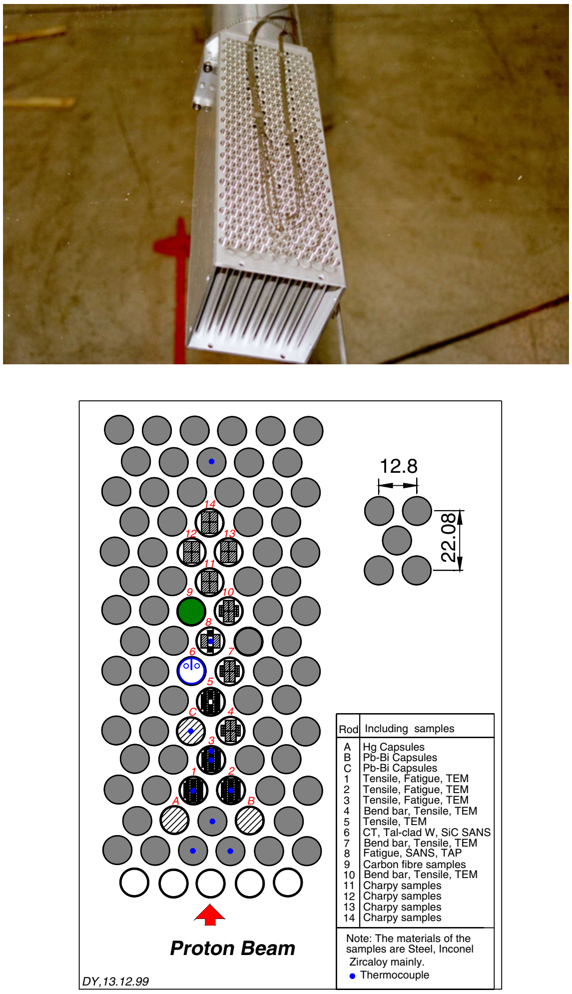

Fig. 5. The rod array of the SINQ solid lead (cannelloni-type) Target 4, along with a sketch illustrating the arrangement of the 17 STIP specimen rods. Temperatures during irradiation are monitored with a total of 10 thermocouples.

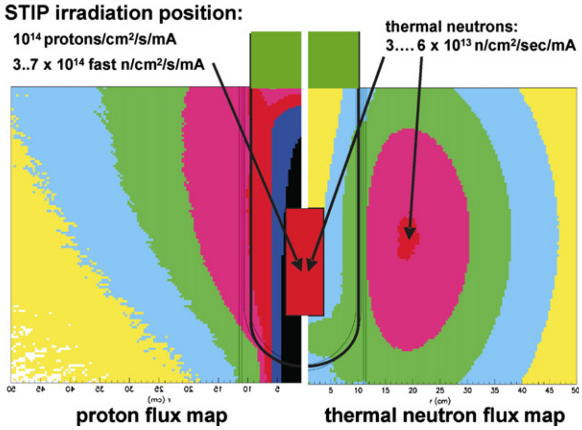

Fig. 6. Flux distribution maps of protons (left) and thermal neutrons (right) for the Target 4 as obtained from MCNPX calculations. Each step in shading represents one decade in flux for the protons, and 1/10th of a decade for the neutrons.

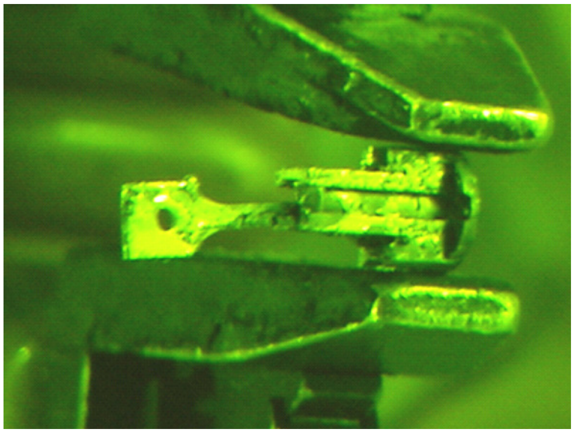

Fig. 7. Martensitic steel (9Cr2WTaV) tensile specimen and stressed capsule irradiated in LBE to 12 dpa in a temperature range of about 210–250 °C. Electron Probe Microanalysis indicates that the LBE corrosion and embrittlement effects are not severe under such irradiation conditions.

the window in the area of the beam entrance is 1.5 mm. After having received a dose of approximately 6.8 dpa in the centre during the scheduled irradiation period, the project partners intend to investigate material properties in an extended post-irradiation examination program, with special emphasis on the beam entrance area of the window.

5. Neutron irradiation rabbit system

SINQ houses a rabbit system for neutron irradiation of materials inserted into the beam tube of Sector 60. Two pairs of rabbit system tubes reach into the volume of the moderator vessel, one to a position closest to the beam tube nose near the target and the other one to a position at a distance of about 80 cm from the beam tube nose. Fig. 9 shows a horizontal cut through the configuration of spallation target and beamport insert housing the rabbit system tubes, and an isometric view of the target and tube noses. Fig. 10 gives an impression of the rabbit system capsules.

The one pair of tubes closest to the target (PNA) is mainly used for isotope production (flux of predominantly thermal neutrons: $2.0 \times 10^{13}/cm^2/s/mA$). Typical nuclides produced are ^{24}Na, ^{82}Br, ^{42}K, $^{175/181}Hf$, ^{131}I, ^{60}Co, ^{198}Au. The second pair at the lower flux position (NAA) is best suited for activation analysis (flux of predominantly thermal

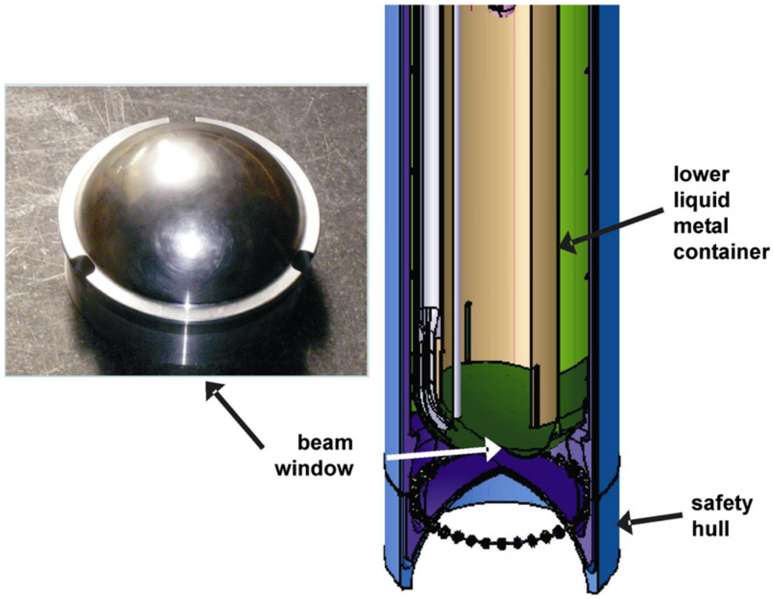

Fig. 8. The well-examined beam window is welded onto the MEGAPIE system as illustrated on the schematic.

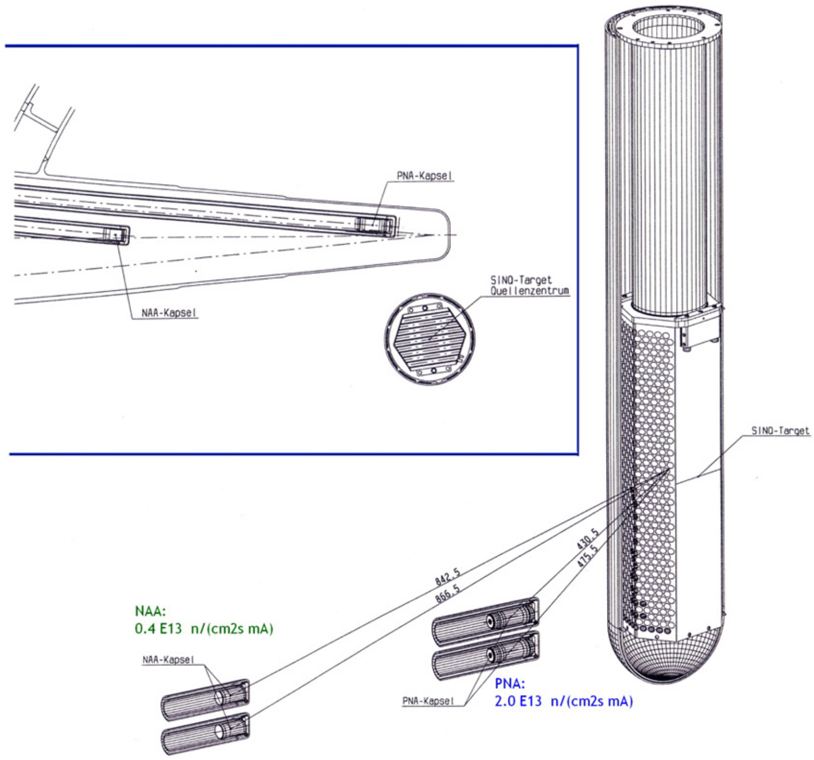

Fig. 9. Neutron irradiation rabbit system at SINQ. Top left: horizontal cut through the configuration of spallation target and beamport insert housing the PNA and NAA rabbit system tubes. Right and below: isometric view of the target and tube noses.

neutrons: 0.4×10^{13}/cm^2/s/mA). Both facilities are actively used, although not overbooked, and will be maintained in the future, with upgrades if required for a dedicated user service.

Fig. 10. Rabbit system irradiation capsules, made of thin-walled aluminium for PNA and of polyethylene for NAA.

One of the specialties of having such irradiation positions at a spallation source is the presence of a certain fraction of high-energy neutrons, which are not present in a reactor. With that our facility allows to study threshold reactions in isotope production and activation which otherwise is not accessible. So far, however, this opportunity has not been deeply exploited.

6. Conclusion

The primary purpose of the Swiss proton accelerator complex at PSI, which combines a 72 MeV and a 590 MeV proton cyclotron and the spallation neutron source SINQ, is the user service for particle physics, muon spectroscopy and neutron scattering and imaging. In addition, several irradiation facilities are operated for investigation of materials behavior under high-dose conditions as well as for neutron activation analysis and isotope production. In LiSoR, connected to the 72 MeV proton accelerator Injector 1, steel samples are irradiated while being in contact with flowing lead–bismuth-eutectic at elevated temperatures and under tensile stress. In the spallation neutron source SINQ, the STIP program allows materials irradiation under realistic spallation conditions, i.e. in a mixed spectrum of 570 MeV protons and spallation neutrons. Within the frame of this program hundreds of samples, mainly austenitic and ferritic–martensitic steels like 316L, T91 or F82H, were irradiated to doses up to 20 dpa. In addition, steel samples in contact with liquid Hg and liquid LBE were part of the program. MEGAPIE, a liquid metal target employing LBE, operated in SINQ during the second half of 2006, can be taken as a materials irradiation facility on its own. Adjacent to the target position, SINQ houses a neutron irradiation rabbit system serving activation analysis and isotope production.

In view of the urgent demand for appropriate materials irradiation facilities from fission and fusion reactor developments, from spallation sources and neutrino factories, the irradiation facilities available at PSI can significantly contribute to dedicated irradiation programs. Although they cannot satisfy all requirements, which are generally diverging, some basic experiences can be gained on materials behavior in high-dose irradiation environments for safety assessments and lifetime predictions. Improvements are under consideration aiming to even better comply with the special needs of the international irradiation community.

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