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Materials related R&D work for the ESS target stations

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Abstract

Current planning for the European Spallation Neutron Source Project (ESS) provides for two target stations of 5 MW beam power each: a long pulse station (2 ms, $16^2/3$ Hz) and a short pulse station (1.4 μs, 50 Hz). In both target stations liquid mercury is the reference target material. Materials related R&D work concentrates on:

- The effects of the high pulsed power input into the short pulse target (stress pulses, pitting, high cycle fatigue) and their mitigation (with emphasis on the introduction of bubbles of non-condensable gas in the liquid metal).
- The effects of radiation on the mechanical properties of structural target components (hardening and embrittlement).
- Investigations of novel cryogenic moderator materials (ice and solid organic compounds) under fast neutron irradiation.

Most of these efforts are carried out together with other projects within international collaborations such as ASTE, JESSICA, LiSoR, STIP, URAM and others.

1. Introduction and brief general information on the ESS Project

Many vital components in or near the targets of high power spallation sources are subject to heavy loads which may determine their integrity and lifetime. The main cause for these loads is the high power density dissipated and the radiation level generated in the materials of a target and its surrounding structures. This refers to both, peak value in the pulse and time-average value. Effects due to the pulsed power input include stress waves, pitting and fatigue whereas the fast accumulation of radiation damage and (possibly) corrosion effects are caused by the high average beam input. Materials issues have been and are therefore an important part of the R&D programs for the target stations of spallation sources in the MW beam power class such as SINQ, SNS and JSNS and, of course in particular for the most ambitious project ESS with an anticipated beam power of 5 MW for each of its two targets.

Before addressing the topic of this review we give some general information on the ESS Project. At present it is in its baselining phase aiming at the delivery of a final report which can serve as the basis for a political decision on the realization of the project. This decision is anticipated in early 2004. Fig. 1 shows an artist's conception of the reference layout of ESS, illustrating its main components: In broadest terms, the facility will consist of two ion sources feeding into a linear accelerator (Linac) with 10 MW beam power and a final particle energy of 1.334 GeV. Half of the beam power will be directed into the long pulse target station (LPTS), the other half will be compressed into μs-pulses in two rings (one lying on top of the other) before entering the short pulse target station (SPTS). Fig. 2 provides a list of parameters of the long and short pulse target stations, respectively, together with a sketch of the pulse sequence required at the end of the Linac. For a detailed description of the reference design of the facility we refer to the recently published ESS Technical Report [1].

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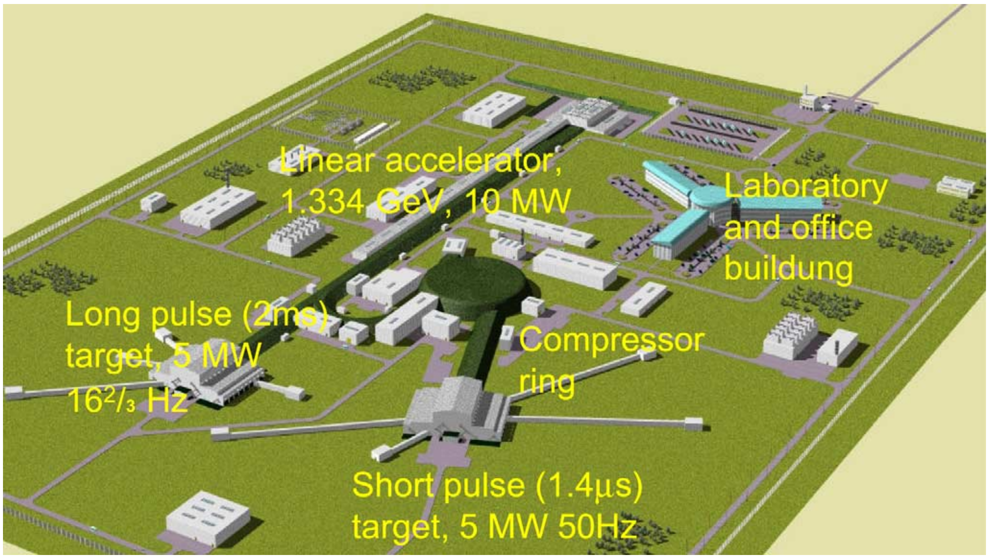

Fig. 1. Perspective view of the schematic layout of the ESS – a 2 × 5 MW spallation source project.

Two target stations	**SP, Short Pulse**	**LP, Long Pulse**
Beam power	5 MW	5 MW
Energy of protons	1.334 GeV	1.334 GeV
Time structure of proton pulse	2 x 0.6µs	2.0 ms
Energy content of proton pulses	100 kJ	300 kJ
Repetition rate	50 Hz	16 $^{2}/_{3}$ Hz
Proton beam footprint at target	6 x 20 cm^2	6 x 20 cm^2
Target type	flowing mercury	flowing mercury
Injection	horizontal	horizontal
Number of moderators (viewed faces)	2 (4)	2 (4)
Average thermal flux	3.1 x 10^{14} n/cm^2s	3.1 x 10^{14} n/cm^2s
Peak thermal neutron flux	1.3 x 10^{17}n/cm^2s	1.0 x 10^{16}n/cm^2s
Effective decay time of flux	150µs	150µs

Required accelerator pulse sequence

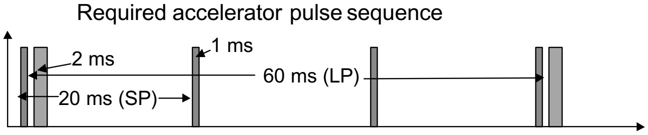

Fig. 2. Short and long pulse target parameters and accelerator pulse sequence for achieving the required pulse structures.

Fig. 3 shows an elevation of the target buildings which are almost identical for the LPTS and SPTS, apart from the types and geometries of the moderators and beam lines. For both stations a liquid mercury target was selected in preference to a water-cooled solid target for several reasons. Some advantages include higher neutron production, better heat removal capability, reduced specific radioactivity and afterheat, and the absence of radiation damage in the target material itself. A three-dimensional view of the reference design of the 5 MW ESS mercury target with its surrounding structures is shown in Fig. 4. The materials in this region experience the highest loads in the facility and their performance is crucial for the successful operation of the ESS. Currently clad lead rods are foreseen as reflector material but other options are being studied.

Critical components include the mercury container, the safety hull, and the proton beam window. The candidate materials for these parts are martensitic and austenitic steels and AlMg3 alloy. Special attention must be paid to the mercury container which is not only subject to intense fluxes of high energy protons and neutrons but also has to withstand high pulsed stresses

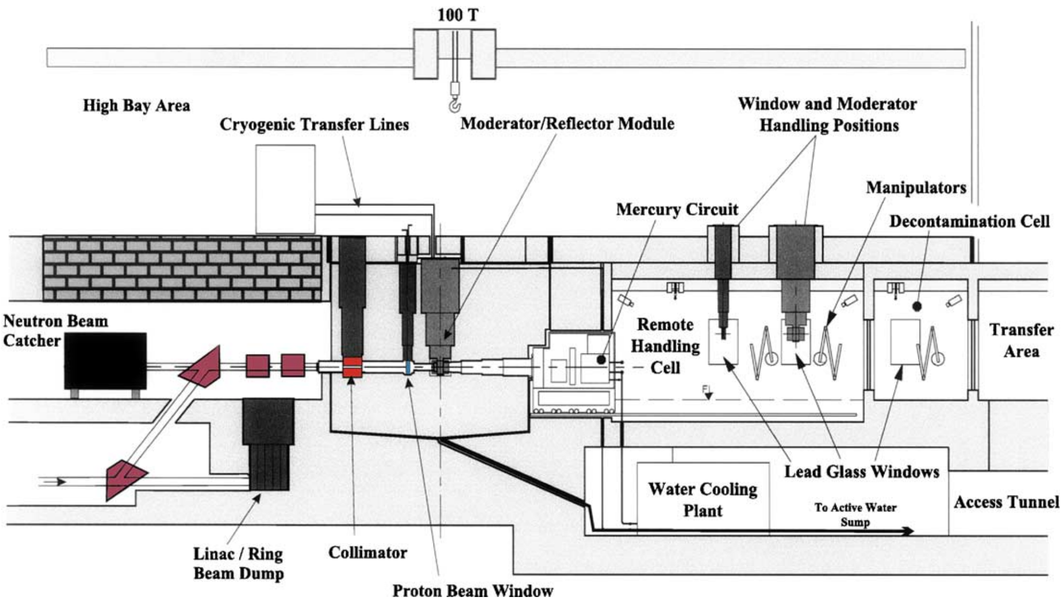

Fig. 3. Elevation of the overall layout of the target stations.

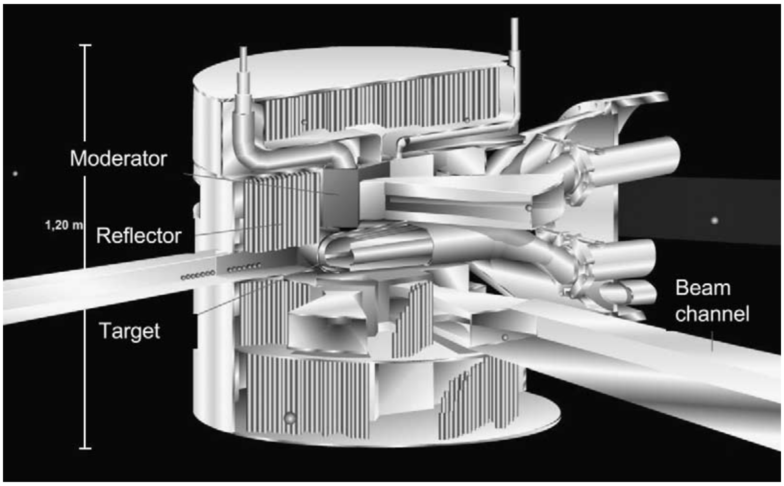

Fig. 4. Sectional view of the ESS target–moderator–reflector systems.

due to pressure waves induced in the liquid mercury. The recently observed 'pitting' is probably connected with these pressure waves. Joint investigations of the projects SNS, ESS and JSNS are under way to try to understand this phenomenon and to mitigate its effects (see Chapter 2). Concerning radiation damage in the structural materials mentioned, a large data base exists for other nuclear environments. However, although the spallation conditions differ from the fission and fusion cases, the main effects of irradiation are well established through results obtained so far from extensive investigations of specimens extracted from spent targets of operating medium power spallation sources (LANSCE, ISIS, SINQ). They do not show any unexpected new qualitative effects of irradiation on the mechanical properties of structural materials. Together with data from dedicated irradiation experiments in SINQ (STIP Program) and LANSCE, it should be possible to select the optimum materials for the target structures of the ESS in the near future. In Chapter 3 we give some selected examples of recent results on R&D efforts in this field. For a more detailed and complete description we refer to the pertinent contributions of the ESS participants to this workshop.

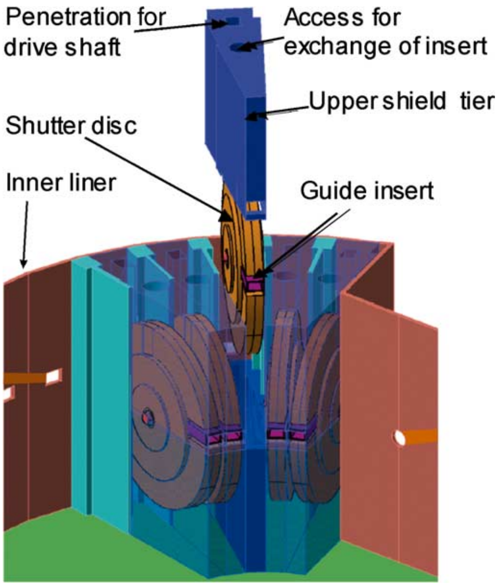

Fig. 5. Arrangement of shutters providing for a small angular separation (11°) between beam lines and a short distance (≈1.6 m) between guides and moderator.

The situation is less clear for a different class of materials in the moderator–reflector-shielding block region, namely novel cold moderator substances (as, e.g. solid methane) which promise superior performance over liquid hydrogen but suffer from severe radiation damage effects which makes their residence time in the moderator volume undesirably short. Investigations of these phenomena and attempts to overcome the problem will be briefly addressed in Chapter 4.

There are other materials-related issues in the ESS target station which will however not be addressed in this short review. An example is the question how long the supermirror inserts in the beam shutters can stand the radiation field which will be rather intense because in the present reference design these inserts are planned to begin at a distance of only 1.6 m from the moderators (Fig. 5).

Recently discussions have been initiated between the European Fusion Community and the ESS Project to evaluate opportunities of incorporating a fusion materials irradiation facility into the ESS target station. Preliminary neutronics and thermal hydraulic calculations together with considerations of the engineering feasibility suggest that this might be a realistic option (Chapter 5).

2. Stress pulses and pitting

For the short pulse target the energy content of a proton pulse entering the mercury every 20 ms is 100 kJ, nearly 60 of which will be deposited as heat in a few litres of liquid metal within 1 μs. As a consequence, the mercury will heat up rapidly and try to expand. Since the low compressibility of the surrounding liquid will hinder this expansion, pressure will build up almost instantaneously and a pressure wave will propagate through the liquid. Impinging on the steel container, it will induce tensile and shear stresses in the wall. Computer calculations with different codes predict stress amplitudes which are close or even above the design stresses of the candidate materials for the mercury container. An example is given in Fig. 6 showing the distribution of the maximum principal stresses and strains (the latter amplified by a factor of 100) in the ESS mercury container 40 μs after the energy input. This is the point in time, when the stress in the container reaches its maximum value. In the centre of the beam entrance region a stress amplitude of almost 400 MPa is obtained for a wall thickness of 1.5 mm.

A comparison of different codes used in the calculation showed good agreement (Fig. 7). The codes also seem to be able to reproduce the time dependence of the initial build-up of the stresses, as a comparison with first experimental results given in Fig. 8 shows. However, a reliable estimate of the magnitude of the stresses is difficult because they depend on assumptions about the behaviour of liquid Hg under tensile stresses. The question of the magnitude of tensile stress liquid metals can sustain has recently gained increased attention after local erosion damage was discovered in steel surfaces in contact with a heavy liquid metal in which pressure waves had been induced by pulsed mechanical or radiation induced loads. Indications are that this pitting [15] (Fig. 9) is caused by the collapse of cavities formed during the tensile phase of the pressure waves.

A possible way to circumvent both the high pulsed stresses and the pitting of the mercury container walls may be to maintain a small concentration of helium bubbles in the beam interaction zone. Their compression should initially accommodate the expansion of the liquid and thereby lower the amplitude of the pressure wave. Calculations for helium bubbles in mercury [3,4] indicate that the maximum stress can be lowered by more than two orders of magnitude as compared to the gas free case (Fig. 10). An experimental verification of this prediction is needed, in particular with respect to the practicality and dynamic response of such a two-phase system. Because of the many advantageous features of the liquid mercury target concept, irradiation experiments at LANSCE (LANL) and AGS (BNL) designed to clarify those issues are pursued with high priority within the ASTE collaboration between the ESS, SNS and JSNS projects. First results are anticipated in the fall of this year and there is hope that they will support a decision on the target concepts to be pursued further for ESS and SNS.

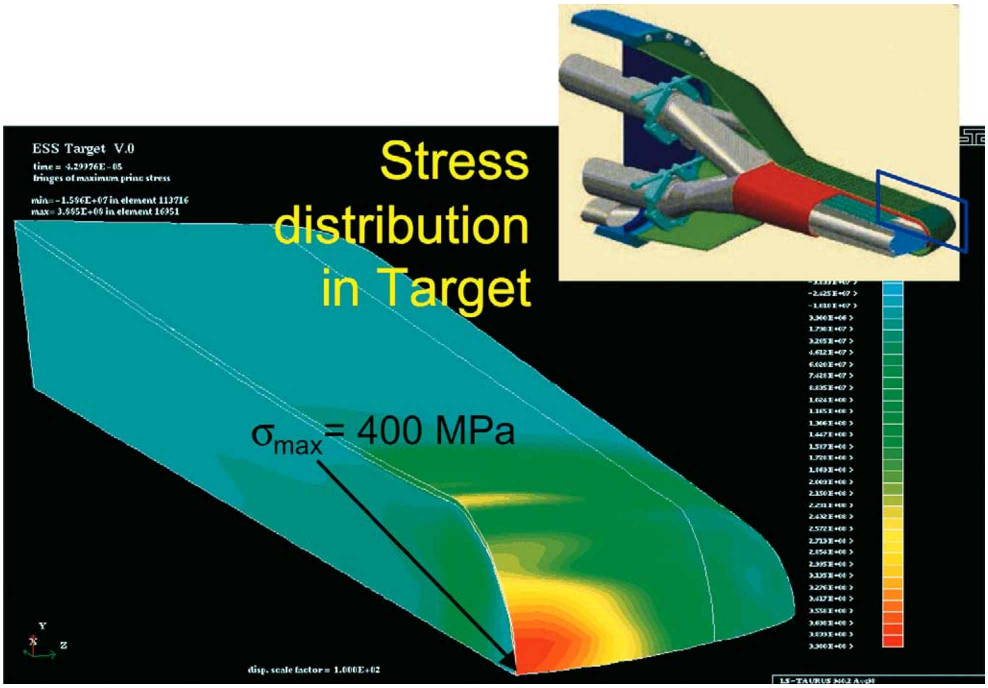

Fig. 6. Distribution of maximum principal stress in the mercury container of the short pulse target at a time of 40 μs after the 100 kJ beam energy input, when the stress in the window region is expected to reach its maximum value. Only one quarter of the target (see insert) is shown. Also given are the deformations of the container walls (amplified by a factor of 100) [2].

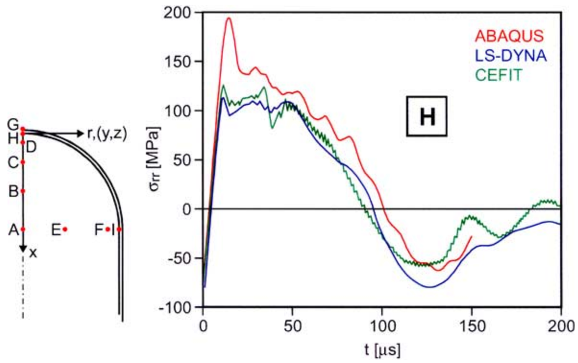

Fig. 7. Comparison of different codes used for computing the stresses as a function of the time after introducing a short energy pulse into the mercury. For the example shown, the cylindrical target used in the ASTE experiments has been chosen. The diagram shows the computed stresses in the inner surface of the container wall at the beam axis (position H in the insert) [13].

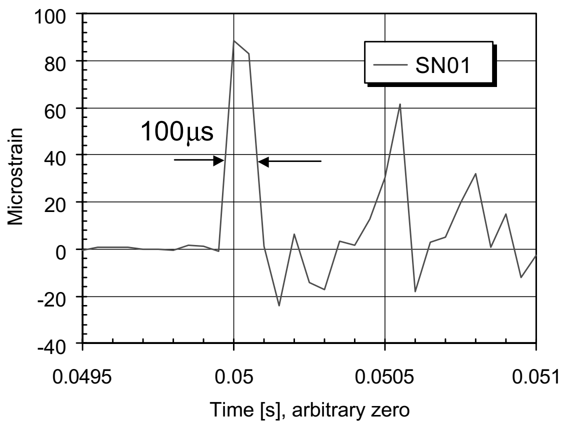

Fig. 8. Strain at position G (see insert of Fig. 7) measured after a proton pulse delivered by the AGS accelerator at Brookhaven National Laboratory (ASTE collaboration).

Another topic related to the fast pulsing is the question whether conventional materials parameters such as yield stress, uniform strain, fatigue endurance limit, etc., are suitable to describe the behaviour of materials strained with the extremely high rates of several 1000% per second as experienced by the components in the SPTS. This issue will be investigated by experiments in a special high cycle fatigue machine driven by a piezo-crystal (Fig. 11). Its design and construction has recently been finished and first test runs were successful. The device will accept reference specimens and specimens irradiated in SINQ within the STIP Program (see next chapter).

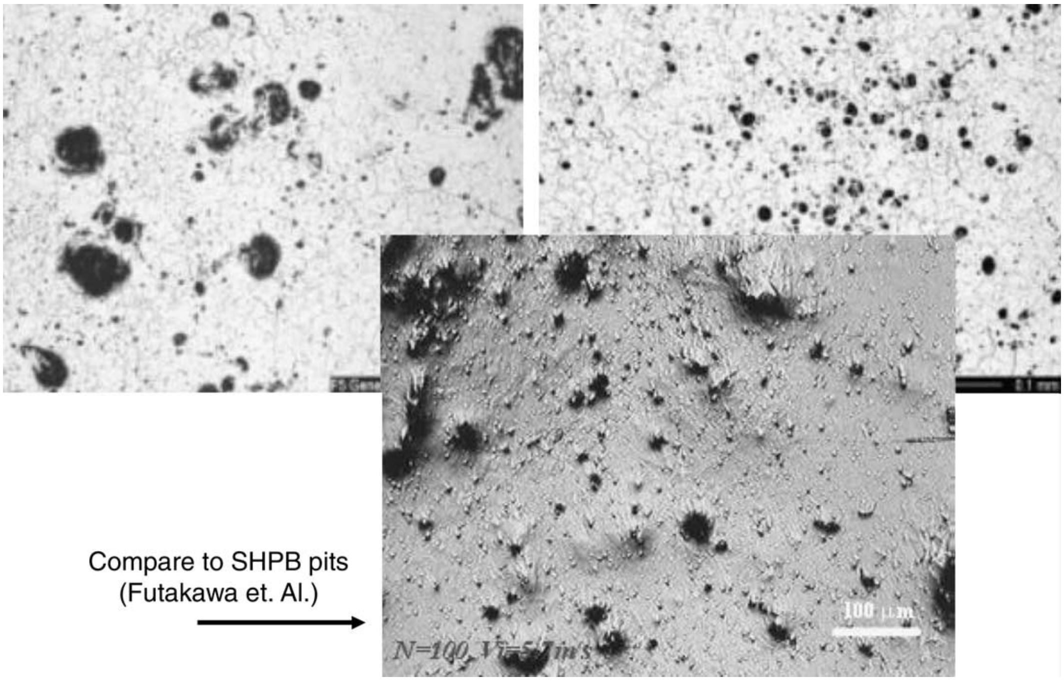

Fig. 9. Pits on the surface of a solution annealed 316L specimen in contact with liquid mercury after 200 proton pulses in WNR irradiation experiments at Los Alamos National Laboratory [14]. The energy density of the short pulses corresponded to SNS or ESS targets operated with a 3 MW time-averaged beam power. The damage is similar to that produced by mechanical shock experiments performed at JAERI in Japan (lower photograph [15]).

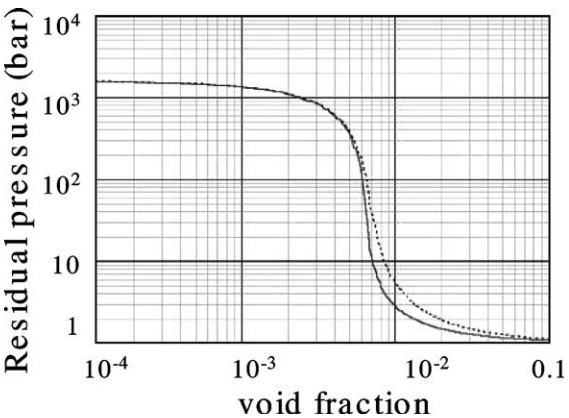

Fig. 10. Predicted reduction of the pressure pulse induced in mercury by the presence of small gas bubbles in the liquid metal. Already a void fraction of less than 1% reduces the pressure amplitude by more than two orders of magnitude [4].

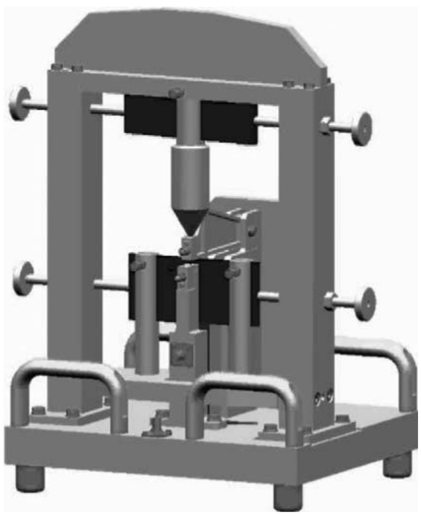

Fig. 11. High frequency fatigue machine capable of loading reference and irradiated specimens with strain rates typical for structures in short pulse targets (up to 50 s^{-1}). First successful tests have been performed at the Forschungszentrum Juelich.

3. Radiation damage

In addition to the mechanical and thermal loads caused mainly by the pulsed energy deposition, the structural materials of high-power spallation sources will experience radiation damage by the high energy protons and neutrons. The resulting changes in the mechanical properties will strongly affect the lifetime of the respective components. Based on ample experience in the development of fission and fusion materials it can be expected that hardening and low temperature embrittlement should be the main effects in the temperature range of room temperature to 250 °C relevant for liquid mercury targets. It is, however, not possible to quantitatively transfer fission and fusion data to the spallation case which is characterized by extremely large helium/hydrogen to dpa ratios and high primary knock-on energies. Irradiations and tests under conditions typical for spallation targets are thus indispensable.

The fastest way to start up a spallation materials data base appeared to be the investigation of specimens obtained from spent targets of already operating medium-power sources. Beginning in 1996, we therefore collected spent target components from LANSCE (Los Alamos National Laboratory) and ISIS (Rutherford-Appleton Laboratory) and a window prototype irradiated for PSI at LANSCE. The available materials included four different classes: an austenitic stainless steel (AISI 304L), a nickel–base alloy (IN 718), a martensitic steel (DIN 1.4926) and a refractory metal (tantalum). The specimen preparation and the measurements were mainly performed in the Hot Cells of the Forschungszentrum Juelich. Several types of mechanical tests including hardness, bending and tensile tests were conducted. The fracture modes were identified by SEM and the radiation-induced changes in the microstructure by TEM. This work is almost finished and a summary of the results in given in the contribution [5] to this workshop. Here we show only two illustrative examples (Figs. 12 and 13) and briefly mention the main conclusion of these investigations: Although all materials show strong irradiation-induced hardening and severe embrittlement, they maintain ductility and toughness values up to 10 dpa which should ensure a safe and reliable function of the respective components up to a service time of about two months full power operation in ESS. The only material for which specimens irradiated to higher dose were available is IN 718, which showed totally brittle fracture in the elastic regime when irradiated to 20 dpa [6] while still retaining 2% uniform strain at 10 dpa. This clearly demonstrates the need to obtain higher irradiation doses on the other materials. Such specimens should become available soon, when the SINQ target is opened, which was in service during 2000 and 2001 (STIP-II irradiation, see below). While results obtained from spent

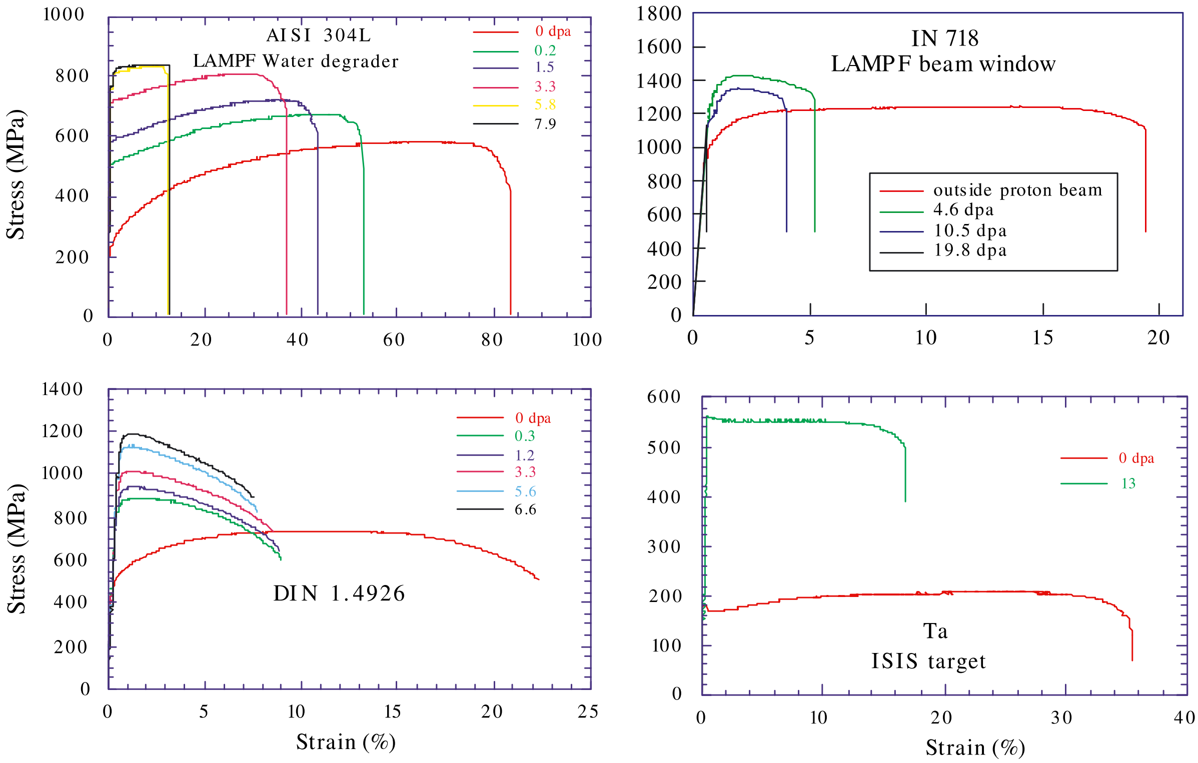

Fig. 12. Examples of stress–strain curves obtained from tensile tests at room temperature on specimens prepared from spent targets of LANSCE and ISIS. For details see contribution [5] to this volume.

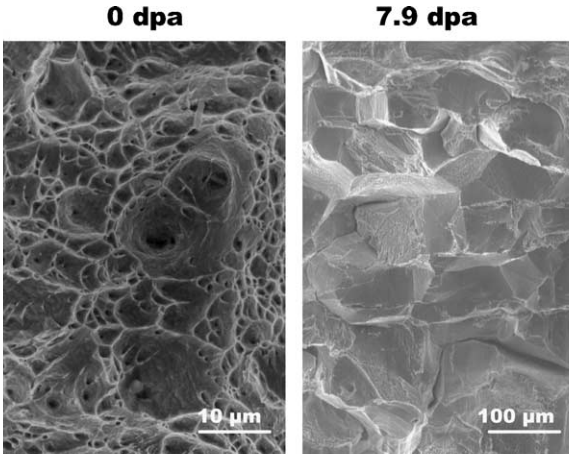

Fig. 13. Fracture surfaces of 304L specimens tensile tested at room temperature. The change from ductile transgranular failure of the reference material to brittle intergranular fracture after irradiation to 8 dpa is evident [16].

targets have the advantage that they reflect the actual service conditions of target components quite realistically (changing power levels, i.e. varying temperatures and dose rates, short and long down times), the complex irradiation history is largely unknown. This makes the allocation of the data to a set of parameters virtually impossible. Moreover, the varying conditions may mask effects which occur only in a narrow range of environmental parameters. The investigations on spent targets must therefore be supplemented by well documented irradiation experiments on standardized specimens. This is to a large extent achieved in the international collaboration STIP (SINQ Target Irradiation Programme) started in 1998 [7].

In this effort several of the target rods in the high flux region of the SINQ target are replaced by instrumented tubes filled with a large number of miniaturized specimens. At present, SINQ is the most powerful operating spallation source, achieving a displacement dose of more than 20 dpa during a typical target service time of about 1.5 full power years. In the first irradiation campaign in 1998/1999 (STIP I), about 1500 specimens of different geometries and made from different candidate materials were irradiated. Late in 2000 they were shipped to the different STIP partners for post-irradiation examination. First results have been published [8]. A second campaign was started in March 2000 and finished in December 2001 (STIP II), reaching a maximum dose of 22 dpa and more than 2000 appm helium in steel specimens. Here some sample rods contained specimens immersed in Hg and PbBi (Fig. 14) to study possible effects of irradiation on liquid metal corrosion and embrittlement, albeit in stagnant liquid metal only. The specimens for STIP III have been prepared and were loaded into the new target for operation in 2002 and 2003. Details on the number and type of specimens and their environmental and irradiation parameters in the different campaigns can be found in Ref. [9].

The post-irradiation examinations of specimens from spent spallation targets and from STIP are supplemented by simulation experiments addressing special topics. The influence of very high helium concentrations on the low temperature mechanical properties is studied by tensile tests and TEM of specimens implanted with helium at the Juelich CV-28 Cyclotron [10]. Another cyclotron at PSI serves as the particle source for the LiSoR (Liquid–Solid-Reactions) Experiment where possible interfacial effects between structural materials and liquid metals will be studied under realistic conditions, i.e. the simultaneous action of irradiation,

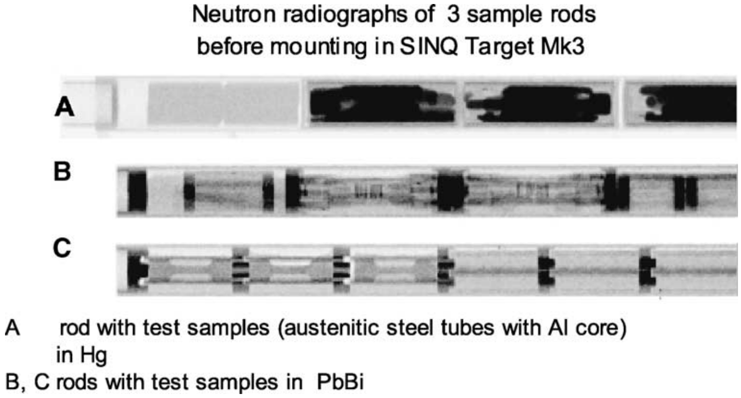

Fig. 14. Neutron radiographs of three sample rods before mounting into the SINQ Target Mk 3 (STIP collaboration [9]).

mechanical stress and flowing liquid. LiSoR started to operate with flowing liquid PbBi in July 2002.

The above listing of activities shows that a good deal of effort goes into the field of radiation damage in structural materials of spallation targets. No unexpected effects have been found for spallation specific irradiation conditions. Together with the results of our partners PSI, ORNL and LANL, the described effort will lead to an extensive data base which should allow (a) the selection of the optimum structural materials for the ESS targets in due time and (b) a reliable prediction of the life times, i.e. the service periods, of critical target components.

4. Novel materials for cold moderators

At present virtually all cryogenic moderators in operating neutron sources use liquid – or supercritical – hydrogen as the moderator substance. This material shows attractive properties and the technology is well established. On the other hand it has been known for a long time that methane in condensed form is, in principle, a much better moderator material, because it has (a) a significantly higher hydrogen density than liquid hydrogen and (b) low-lying modes in the energy region 1–3 meV (5–15 K) which the neutrons can excite. In this range solid methane gives three times more neutrons than liquid hydrogen in the same geometric configuration (Fig. 15).

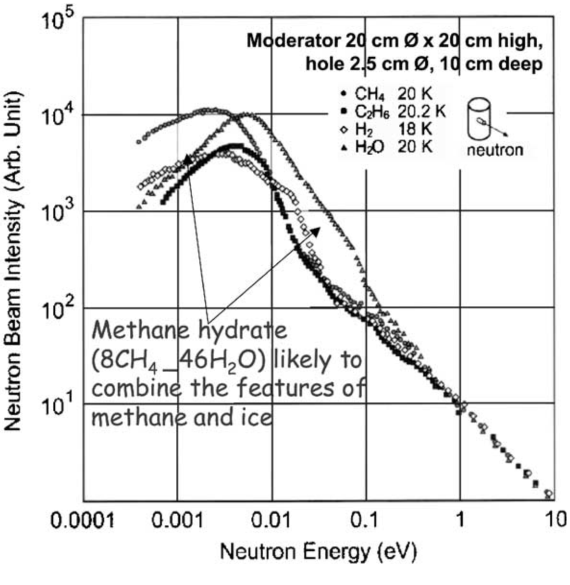

Fig. 15. Comparison of neutron intensities emitted by a cold moderator filled with different candidate materials. Ice gives up to 5 times more thermal neutrons than any other cold moderator, solid methane gives 3 times more cold neutrons than the present reference of supercritical (liquid) hydrogen [17].

Unfortunately the use of solid methane is not straight-forward in practice because of its low thermal conductivity and its extreme vulnerability to radiation damage. Its molecules are destroyed by the fast neutron and γ irradiation, leading (a) to accumulation of hydrogen and eventual spontaneous release in a catastrophic manner and (b) to the formation of waxy residues that tend to stick to the container walls and pipe work. The first problem is common to other promising cold moderator substances such as water ice which yields up to five times more neutrons at energies between 5 and 100 meV than any other cold moderator (Fig. 15).

It turned out that methane hydrate, i.e. ice with methane molecules trapped in cages of the crystal lattice, combines the high density of states of ice above 50 meV with those of methane below 30 meV and features the highest hydrogen density. It may therefore be speculated that methane hydrate would make the ideal cryogenic moderator material, if a system can be devised that allows heat removal on the one hand and periodic annealing of the radiation damage on the other [11].

In view of these promising prospects R&D efforts are being spent to (1) verify these predictions and (2) develop systems that use small pellets of either one of the three materials which would be continuously transported through the moderator vessel and would be cooled either by supercritical hydrogen or liquid helium. At present, the activities concentrate on

- the production of pellets of methane and methane hydrate,
- irradiation experiments of the novel materials in the IBR-2 reactor at JINR Dubna (URAM collaboration [18]),
- development of scattering kernels for methane hydrate,
- verification of the performance expectations in JESSICA,
- the development of transport and cooling systems for the pellets.

The work is pursued within the AcoM collaboration and with a number of other partners.

Irradiation experiments carried out at the IBR-2 reactor in Dubna (Russia) under a joint programme called URAM-2 clearly showed that trapping of radiolytically generated hydrogen is a common feature of all molecular moderator materials examined. Fig. 16 shows temperature recordings inside the moderator, in the copper container and in the helium coolant. It can be seen that, as the temperature is allowed to rise gently after the irradiation, a sudden jump occurs, which indicates the spontaneous exothermal recombination of radicals after the mobility of the hydrogen has reached

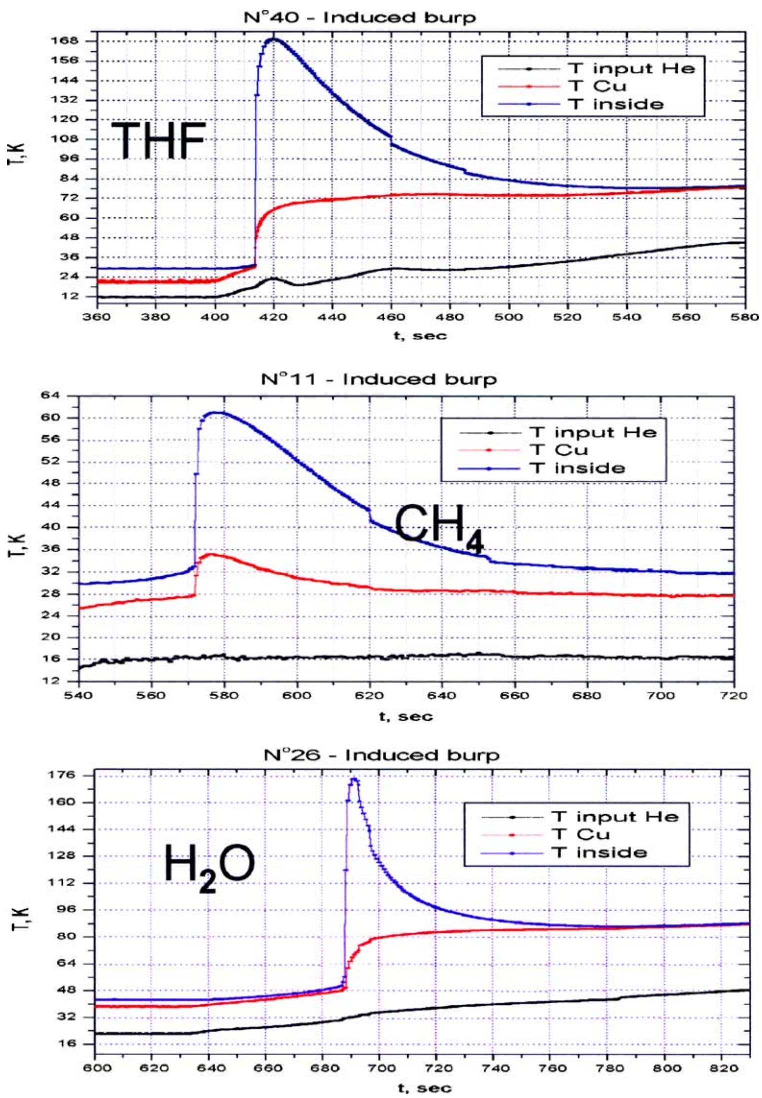

Fig. 16. Sudden release of stored energy in solid cold moderators ('burping') during irradiation with fast neutrons leading to a drastic temperature increase in the moderator substance [19].

the corresponding threshold. Tetrahydrofurane (THF) was used as a surrogate clathrate for methane hydrate because it is much easier to manufacture. The curves also confirm that water ice has a thermal conductivity which is much higher than either methane or THF, since it recovers from the temperature jump much faster.

The Jülich Experimental Spallation Target Setup in COSY Area (JESSICA) is a 1:1 replica of the ESS target/moderator/reflector unit reference design, which uses the 2.3 GeV Cooler Synchrotron COSY in the Forschungszentrum Jülich as its pulsed proton source. The JESSICA setup consists of a target container filled with (stagnant) mercury and surrounded by a reflector composed of lead cylinders. It allows changes in the sizes and positions of the volumes available for different types of moderators. This flexibility together with the handling devices and cryogenic equipment installed in the experimental area make JESSICA an ideal test bed for the optimisation of the target/moderator/reflector design and for the verification of codes used to predict the neutron performance of ESS target stations. This is illustrated in Fig. 17, which shows a comparison between the spectrum of neutrons produced by a room temperature water moderator in JESSICA to the results of calculations with the MCNPX code. Experiments with cold moderators are under way and first results clearly show the changes in pulse width and intensity relative to the room temperature results.

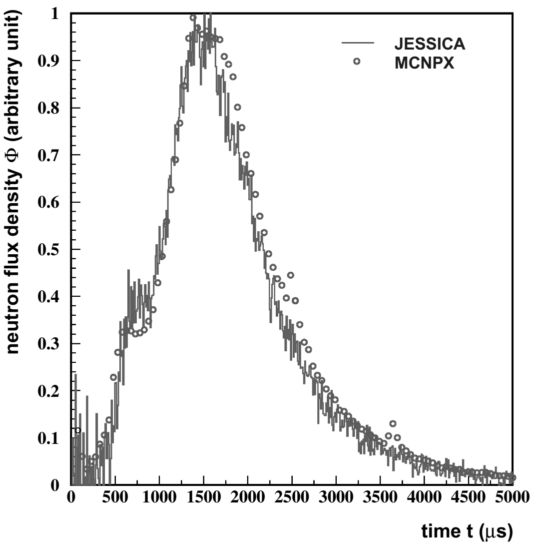

Fig. 17. Comparison of measured and calculated neutron flux densities provides by moderators. The example gives the measured thermal spectrum of a water moderator in the JESSICA facility and shows that it can be accurately simulated by the MCNPX code [20].

5. An option for a materials irradiation facility

Fusion materials research is impeded by the lack of a prototypic irradiation source. During the past four decades several projects have been proposed but could not be realized until now, mainly because of the high costs for such a single purpose facility. At present the IFMIF (International Fusion Materials Irradiation Facility) is under discussion aiming at start of construction and operation in 2008 and 2016, respectively. For the recently proposed 'fast track towards fusion energy production', it was deemed desirable to 'identify to which extent relevant studies could be done on Neutron Spallation Sources available now and in the foreseeable future in Europe or elsewhere' [12]. Triggered by this request, neutronics calculations and engineering considerations have been initiated to examine the possibility of incorporating a fusion materials test module in an ESS target station. First preliminary results obtained by independent calculations in the research centers at Juelich and Karlsruhe [22] show good agreement and indicate that irradiation parameters relevant for a fusion DEMO reactor can be achieved in irradiation rigs placed in the reflector region just above the target and downstream from the moderator (Fig. 18 and Table 1). An estimate of the production of impurity elements (e.g. Ca, S, P) yielded values in the same order of magnitude for ESS as for the IFMIF case, i.e. concerns about a negative influence of the 'high energy tail' of the spallation neutron spectrum could be dispelled. Also, no principal engineering problems were identified. Therefore the installation of a materials irradiation module in the reflector of an ESS target station might be a useful option, whose pursuit would, however, require further efforts to optimise the design and working out some engineering details.

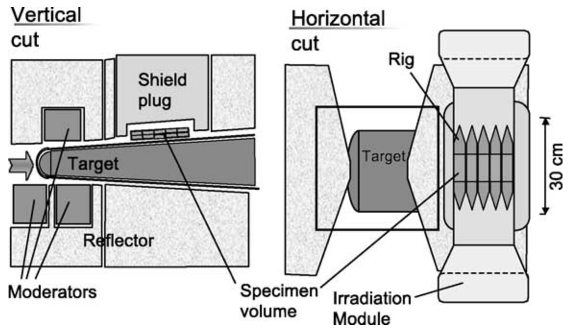

Fig. 18. Possible position of an irradiation module in the reflector of the ESS target station under consideration for fusion materials research. The attainable irradiation parameters are given in Table 1 [21].

Table 1
Irradiation parameters attainable in a fusion materials test module, placed in the reflector of an ESS target station

	ESS A	ESS B	DEMO 2 MW/m^2
Total neutron flux (n/m^2s)	1.0×10^{19} (5×10^{15} p)	5.3×10^{18} (2.3×10^{15}p)	7.1×10^{18}
Displacement rate (dpa/s)	3.6×10^{-7}	1.4×10^{-7}	5.4×10^{-7}
He production rate (appm/dpa)	3.0×10^{-6}	7.1×10^{-7}	5.7×10^{-6}
He/dpa ratio (appm/dpa)	8.5	5.1	11
Displacement per FPY (dpa)	11.2	4.3	17

A is a volume element in the left (upstream) position of the module (see Fig. 18), B is in the right (downstream) position. The corresponding values in the first wall of a DEMO fusion reactor with a wall loading of 2 MW/m^2 are given for comparison [21].

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