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Summary

Summary of the Fourth International Workshop on Spallation Materials Technology (IWSMT-4)

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Abstract

The Fourth International Workshop on Spallation Materials Technology (IWSMT-4) took place on 8–13 October in Schruns, Austria. The present volume contains the proceedings of this workshop. 53 scientists from Europe, USA and Japan presented 44 papers covering a wide range of topics including the status of spallation target R&D activities, effects of radiation damage, hydrogen, helium and other transmutation impurities in materials, particle transport calculations, heavy liquid metal corrosion and compatibility, and materials engineering. The main progress, with an emphasis on materials technology in different projects, namely the US Spallation Neutron Source (SNS), the European Spallation Source (ESS), the Japanese Spallation Neutron Source (JSNS), the Swiss Spallation Neutron Source (SINQ) and the US Accelerator Production of Tritium facility (APT) was overviewed. Progress from the European TECLA and SPIRE programs was described. The achievements of investigations in the last two years on mechanical properties and microstructures of materials under high-energy proton (spallation), neutron and low energy ion irradiations were reported. The latest results from studies on corrosion and embrittlement effects of heavy liquid metals (Hg, Pb–Bi and Pb) were presented. Some critical issues of liquid metal technology related to accelerator driven systems (ADS), e.g. oxygen control and oxygen measurements were discussed.

1. Introduction

For high-power ($\geqslant 1$ MW) spallation targets, liquid metal targets have several potential advantages over water-cooled solid targets, including increased target materials lifetime, better heat removal characteristics and neutronic performance. However, to use such a novel target, there are naturally difficulties and open questions, particularly associated with the structural materials performance under pulsed loads, severe radiation damage and a corrosive environment. This series of workshops is aimed at reporting the latest progress of R&D in different laboratories, discussing open questions and providing technical support for the engineering design of targets for future spallation facilities.

2. Overview of the status of spallation target R&D activities with an emphasis on materials technology

For all the projects, the target R&D program consists of a wide range of both calculational analyses and experimental tests, which cover materials performance, partial transport calculations, and thermal-hydraulic studies. Here, only the activities relevant for materials research are briefly described.

The structural materials in a target, particularly at the proton beam window, are exposed to a severe irradiation, mechanical loads and corrosive environment. The materials performance determines the target safety, availability and lifetime. Therefore, in each of the involved laboratories, there are significant efforts in materials investigations. At Forschungszentrum Jülich (FZJ), the retired components from existing spallation sources (LANSCE and ISIS) have been investigated. Comprehensive results of mechanical tests at room temperature and microstructural studies on steels 304L,

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DIN1.4926, Inconel 718, and pure tantalum have been obtained. In addition, helium effects were studied by cyclotron implantation in martensitic steel F82H and austenitic steel 316L at helium concentrations up to 1 at.%. At Los Alamos National Laboratory (LANL), a large irradiation experiment has been carried out within the Accelerator Production of Tritium (APT) program in collaboration with other national laboratories in the USA. More than 5000 samples, mainly from 316, 304, IN 718-PH, and 9Cr–1Mo were irradiated at 160°C and below to doses up to 12 dpa. Data on tensile properties and fracture toughness have been obtained for all these alloys. At the same time, the microstructures in some materials also have been investigated, and moreover, the hydrogen and helium concentrations were analyzed. At Oak Ridge National Laboratory (ORNL), steels 316LN, 9Cr–1Mo and IN 718 in the PH and SA conditions were irradiated with neutrons and mechanical properties were compared with those from the same materials irradiated with protons. These materials were also studied by triple-ion-beam irradiation with Fe, He and H with spallation relevant He/dpa and H/dpa ratios. To investigate helium and hydrogen effects, steels were implanted up to 20 at.% with He alone, H alone or He and H simultaneously. Hardness tests and TEM observations have been performed on these samples. Numerous experiments on mercury corrosion and compatibility were done in stagnant mercury. Some of these were carried out with concurrent proton irradiation at about 2 MeV. Similar concurrent proton irradiations with flowing coolant were also carried out to investigate irradiation assisted stress corrosion cracking phenomena relevant to spallation targets. At the Paul Scherrer Institut (PSI), the first irradiation experiment in the SINQ target (STIP-I) was finished during 1998 and 1999, and more than 1500 samples were irradiated up to about 12 dpa in the temperature range 80–400°C. The second irradiation (STIP-II) was started in March 2000 and will be finished by the end of 2001. There, more than 2100 samples are being irradiated in the temperature range 80–480°C. The maximum dose in steel samples will be up to 20 dpa or more. To study liquid metal corrosion under irradiation, the LISOR (liquid–solid reaction) project is being carried out in collaboration with SUBATECH/CNRS, France. A Pb–Bi loop is under preparation and is to be installed at a 72 MeV proton beam line at PSI. Further, a 1 MW pilot experiment for a liquid metal (Pb–Bi) target (MEGAPIE) is under development within a wide international collaboration. Investigations of the materials behaviors under irradiation and corrosion are emphasized in the related R&D program and detailed post-irradiation examinations are planned. At the Japanese Atomic Energy Research Institute (JAERI), tensile and bend fatigue tests on some samples irradiated in STIP will be performed soon.

3. Effects of radiation damage in structural materials

Comprehensive results of mechanical properties and microstructure in 316L, 316LN, 304L, IN 718-PH and 9Cr–1Mo were obtained and presented by a number of authors from the materials irradiated at LANSCE with 800 MeV protons. The samples included those from an APT experiment and from a Spallation Neutron Source (SNS) materials experiment irradiated at the same time at the same location. These were irradiated at 160°C and below. Some data also were obtained from the IN 718-PH or 304L beam windows or water degrader of LANSCE that were irradiated at higher temperatures. The mechanical tests were mainly done at room temperature, with some performed at temperatures up to 160°C. The results demonstrated considerable changes in both mechanical properties and microstructure after irradiation.

3.1. 316L/304L stainless steel

Tensile tests were performed on samples exposed to doses up to 11 dpa. The results showed significant increases in yield stresses and decreases in ductility. Nevertheless, the uniform elongation retained 6% at the maximum dose of 11 dpa. Increasing the test temperature from 20°C to 160°C resulted in decreasing uniform elongation. It was found that radiation hardening and loss in ductility were in agreement with databases for materials irradiated in fission reactors, except for more radiation strengthening at high doses, which may be due to the higher gas contents of the proton irradiated materials. The toughness gradually decreased to less than 120 MPa$\sqrt{m}$ after 5 dpa exposure.

The irradiation-induced microstructures mainly consist of small defect clusters and larger faulted Frank loops. Numerous stacking fault tetrahedra were observed in the samples from the 304L window in a dose range 0.7–7 dpa. In this dose range, the densities of both small clusters and Frank loops were insensitive to the irradiation dose. The size of the small clusters was about 1.6 nm, independent of the dose. The size of Frank loops was more or less proportional to the dose. Due to the saturation of small defect clusters, further hardending at doses above about 0.5 dpa should be mainly attributed to the Frank loops. Helium may have had some contribution at high doses.

The main feature in the deformed samples was the formation of dense twin lamellae and bundles of twin lamellae parallel to the {1 1 1} planes. The width of the twin lamellae was as small as a few of nanometers but that of bundles was as large as hundreds of nanometers. The original existing microstructure (small defect clusters and Frank loops) was almost completely removed in the twin lamellae but small changes occurred outside the lamellae. In spite of the large levels of helium and hydrogen retention, no cavities were observed. The precipitates changed from crystalline to amorphous with increasing dose.

Deformation microstructures observed in 316 LN irradiated at 200°C to various very high-helium concentrations and damage levels with 360 keV He-ions and deformed to 10% strain at room temperature by a disk-bend method show similar features, namely the formation of dense twin lamellae. Some evidence indicated that the presence of helium bubbles imposed an additional restriction on dislocation motion, not only in the matrix but also in the channel bands, raising the hardness beyond the level achievable by displacement damage alone. However, this additional effectiveness of helium pinning was observed at a helium level above 1 at.%.

Another interesting observation was that helium started to be dominant in hardening at concentrations above 1 at.% in steels.

3.2. Alloy IN 718-PH

Tests of APT samples show that the uniform elongation drops to less than 2% after only 0.5 dpa of exposure. The yield stress increases slightly at low doses and then decreases with dose above 1 dpa of exposure. The fracture toughness dropped to less than 80 MPa$\sqrt{m}$ after 2 dpa exposure. For the 718 window of LANSCE, a small crack occurred while in service. Tensile and hardness tests performed on the samples from the window were consistent with the tensile data of APT samples. Inspection of the window revealed that the crack occurred at a spot welded thermocouple.

The microstructures of the samples irradiated to 0.1–13 dpa were studied. These show that superlattice spots, corresponding to the age-hardening precipitate phases γ' and γ'', are lost from the diffraction patterns for alloy 718-PH after only 0.6 dpa. Examination of samples that were neutron irradiated to doses of ~0.1 dpa showed that precipitates were faintly visible in diffraction patterns but were rapidly becoming invisible. It was proposed that the γ' and γ'' first became disordered (at < 0.6 dpa), but remained as solute-rich aggregates that still contributed to the hardness at relatively low dpa levels, and then were gradually dispersed at higher doses. At low doses, the dominant microstructural features were black-spot damage and remnants of the γ' and γ'' phases. The black-spot damage and remains of the precipitates resulted in a net hardening up to 0.6 dpa. At doses between ~0.6 and ~13 dpa, the observable microstructure was dominated by only black-spot damage and larger faulted Frank loops. In this dose range the gradual softening was attributed to mixing-induced redistribution of the solutes from the former γ' and γ'' precipitates as the dose increased.

Again, no cavities or bubbles were observed at doses as high as ~13 dpa although the materials should contain about 2000 appm He at this dose level.

3.3. 9Cr–1Mo tempered martensitic steel

Samples irradiated in the APT program up to 9.3 dpa were tested at temperatures of 50°C, 164°C, 250°C and 400°C. The tests at 250°C and below showed that the yield stress significantly increased while the uniform elongation decreased drastically to less than 2% at only 0.5 dpa. A slightly higher (over 3%) uniform elongation was observed along with a decrease in yield stress when the test temperature was raised to 400°C. The fracture toughness dropped to less than 80 MPa$\sqrt{m}$ after about 4 dpa exposure. The toughness dropped faster for crack growth parallel to the rolling direction than for crack growth perpendicular to the rolling direction.

In addition to the above results for 316L, 316LN, 304L, IN 718-PH and 9Cr–1Mo irradiated at LANSCE, some other interesting results were also shown in the workshop. One of them was from the examination of the ISIS tantalum target, which demonstrated that high-purity Ta possessed a good resistance to radiation damage: after 13 dpa exposure the material still showed more than 15% ductility, while its strength had nearly doubled.

4. Helium and hydrogen measurements

The high-production rate of transmutation impurities, especially helium and hydrogen, is one of the main differences between high-energy proton irradiation and fission/fusion neutron irradiation. For spallation materials investigations, it is thus of special interest to determine both the production and retention of helium and hydrogen precisely. Three papers presented in this workshop show interesting results in this connection.

Production cross-sections for neutrons and charged particles (p, d, t, He^3, He^4) as well as excitation energy distributions have been measured by the NESSI (spell out acronym) collaboration for high-energy (0.8–2.5 GeV) proton-induced spallation reactions. For Fe, the radiation damage cross-section is almost constant for proton energies above about 1 GeV, while He production still increases with proton energy. For heavy metals such as Ta a similar result is found for proton energies above about 3 GeV.

The helium and hydrogen contents in the samples and dosimetry disks irradiated in the APT program have been measured systematically. The results showed that helium generation rates in typical iron-base and nickel-base alloys were insensitive to alloy composition during irradiation with protons in the 500–800 MeV range, especially when concurrent neutron fluxes were relatively small. The helium generation rates of mid-Z elements were measured to be larger by roughly a factor of two than those calculated by LAHET, when the code was optimized to reproduce the p/n ratios characteristic of high-Z materials. Iron-rich alloys had measured helium contents in the 150–160 appm He/dpa range. This indicates that separate physics parameters are required for each range of atomic numbers. The ratio of $^4He/^3He$ generation appeared to be somewhat sensitive to proton energy, increasing as the proton energy increased. Hydrogen generation was roughly independent of composition and proton energy within the 500–800 MeV range, but there were energetic losses of the order of 50%, and diffusional losses that varied somewhat, depending on the solubility of hydrogen and the density and nature of both pre-existing and radiation-induced microstructural sinks. A surprisingly large fraction of the deposited hydrogen appeared to be retained in all alloys tested during irradiation below 100°C, where data from unirradiated materials suggest an almost complete diffusional loss. These retention rates indicate that the currently employed cross-sections for hydrogen generation do not need modification. Hydrogen retention appears to be related to irradiated-induced microstructural evolution.

In another study using cyclotron implantation techniques, the diffusion and retention of hydrogen and helium were derived from permeation and thermal desorption experiments. It was illustrated that some hydrogen was effectively retained in martensitic stainless steels even at elevated temperatures due to trapping at irradiation-induced defects, and confirmed that helium is immobile even at elevated temperatures.

5. Liquid metal compatibility

5.1. Mercury

The corrosion and wetting of steels in Hg were tested at about 300°C either in flowing Hg or in stagnant Hg. In some of the tests, thermal convection loops containing mercury and a variety of 316L, 316LN, and IN 718-PH coupons representing variable surface conditions and heat treatments have been operated continuously for periods up to 5000 h. The maximum temperature in the loops was about 305°C; the minimum temperature was about 240°C. Wetting by Hg was somewhat sporadic and inconsistent, and was generally encouraged by steam cleaning and/or gold-coating of specimens prior to testing as well as by relatively high-exposure temperatures. Interaction of 316L and Hg was observed to generate a porous surface layer substantially depleted of Ni and Cr, which resulted in transformation to ferrite, but the maximum penetration detected for all of the test conditions corresponded to only about 60–70 μm/y, with far less penetration for most exposures. In limited testing, alloy IN 718-PH was found to be more resistant to wetting/attack than 316L. In other tests, detailed surface analyses were performed on samples immersed in stagnant Hg at 300°C up to 5000 h. When oxygen was present in the cover gas, the surface layers consisted of a set of oxides Fe_3O_4, Fe_2O_3 and Cr_2O_3 several hundreds of nanometers thick for martensitic steels F82H and MANET, but only several tens of nanometers thick for austenitic steel 316L. A large amount of red HgO single crystals of sizes up to 1 mm was formed on the surfaces of all the samples.

5.2. Pb and Pb–Bi

The corrosion behavior of both austenitic and martensitic steels in Pb and Pb–Bi in a temperature range from 300°C to 550°C was investigated under a variety of stagnant and flowing conditions. Generally, the austenitic stainless steels examined were quite resistant to Pb and Pb–Bi with negligible interaction at temperatures $\leqslant$ 400°C, and only very modest weight change and film build-up (0–4 μm) up to 3000 h of exposure up to 550°C. In contrast, the martensitic steels exhibited generally more extensive attack. Depending on the specific steel and exposure temperature, significant weight gains and an oxide thickness of 10–50 μm (increasing in a parabolic fashion with time) were generally observed on post-test specimens. Typically, the oxides on steels tended to exhibit at least two 'phases' – an outer layer of magnetite that tended to be somewhat porous and partially penetrated by Pb, and an inner layer described by Fe,Cr_3O_4 as a spinel-type oxide. In some cases, a very thin 'diffusion layer' or 'internal oxidation layer' was also observed at the oxide/steel interface. It was confirmed that precise measurement/control of the oxygen content in the Pb or Pb–Bi is required to control dissolution/oxidation of the containment material. In particular, it was emphasized that hydrogen production in a spallation target might tend to deplete oxygen in the liquid metal, potentially to levels below that required to maintain a stable protective film.

To study the embrittlement effects of Pb and Pb–Bi on martensitic steels, particularly 9Cr–1Mo (T91), tensile tests were performed. It was found that when 9Cr–1Mo was tempered at 500°C (instead of 760°C) to increase its hardness, the material was seriously embrittled by liquid Pb at temperatures between 350°C and 450°C, if the surface of the specimens had been additionally hardened by machining a notch into it. In another study, slow strain rate tensile tests showed that embrittlement occurred for T91 and F82H samples tested in Pb–Bi at 300°C when there was some (uncontrolled) oxygen content in the Pb–Bi.

Since the oxygen content in Pb–Bi was found to be important for both corrosion and embrittlement, it should be precisely measured. Recent efforts lead to the successful development oxygen meters at different laboratories. It was found that electrochemical oxygen meters with reference systems such as Pt/air and In/In_2O_3 could be used as in situ devices for measuring the chemical activity of dissolved oxygen in liquid Pb–Bi. However, the application temperature is limited to 400°C and above.

6. General observations

This workshop was the fourth in a series begun in 1996. The complexion of the meetings has changed progressively from early discussions of R&D plans and examinations of relatively sparse data to the presentation of large volumes of new experimental and calculational data. International collaborations that were planned starting about five years ago are now a reality and are heavily utilized in accomplishing the research.

One significant result of these meetings and the supporting research to spallation source designers is that although there are many materials issues that are gradually being resolved in order to define acceptable operating limits, no deadly problems have been uncovered that would rule out the use of heavy liquid metal spallation targets.