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Tensile tests on MANET II steel in circulating Pb–Bi eutectic

H. Glasbrenner [a,b,*], F. Gröschel [b], T. Kirchner [c]

[a] Forschungszentrum Karlsruhe, P.O. Box 3640, 76021 Karlsruhe, Germany
[b] Paul Scherrer Institut, 5232 Villigen PSI, Switzerland
[c] Ecole des Mines de Nantes, 4 rue Alfred Kastler, 44000 Nantes, France

Abstract

Off-beam tensile tests have been performed on MANET II steel in the eutectic Pb–55.5Bi (LBE) and Ar during commissioning of the LiSoR loop, an experimental liquid metal loop, which was developed to investigate the influence of Pb–Bi on possible structural materials under static load and irradiation. Test temperatures were 180–300 °C. MANET II (11% CrMoVNb steel) exhibits good swelling and creep resistance behaviour under irradiation up to around 500 °C. Good corrosion resistance of this material is expected due to the absence of the element Ni in the steel matrix which has a high solubility in LBE. All specimens showed a ductile fracture in Ar. In LBE a loss of ductility was observed at the test temperatures of 250 and 300 °C in comparison to the Ar samples. SEM analysis of the fracture surface of these specimens revealed a mixed mode, i.e. dimple and brittle fracture and penetration of Pb–Bi along the grain boundaries, which is a typical finding for liquid metal embrittlement.

1. Introduction

Spallation sources, which produce neutrons by means of protons fired on a target consisting of heavy metals, are requested for research in material science and condensed matter physics. Liquid mercury as target material is foreseen to be used in the spallation neutron source (SNS) already under construction at ORNL (USA) [1] and the European spallation source (ESS) which is presently in the development stage [2]. Both spallation sources will operate with a pulsed beam bombarding the target to generate spallation neutrons.

The megawatt pilot experiment (MEGAPIE) under design is based on the liquid lead bismuth eutectic (LBE) alloy as target material. The target will be installed in the existing spallation neutron facility SINQ at PSI. For a future accelerator driven system (ADS) LBE is foreseen likewise as spallation target and coolant due to its favourable physical, chemical and thermodynamic properties, e.g. radiogenic lead (Pb-208) and bismuth have the lowest neutron absorption cross sections of all the other potential liquid coolants.

Special properties are required for the window material in the spallation target environment. A material is needed which exhibits good swelling and creep resistance under irradiation at elevated temperature and which shows a quite good corrosion resistance against LBE. Martensitic steels containing 9–12% Cr and about 1% Mo, have been developed and used successfully as core component materials to high fuel burn-ups in sodium cooled fast breeder reactors in France, Germany and UK. These steels have relatively high strengths at temperatures up to about 550 °C and are resistant to thermal stress development, irradiation-induced void swelling, high temperature grain boundary (helium) embrittlement and irradiation creep. The martensitic stainless steel 9Cr1MoVNb (industrial grade T91) has been selected as candidate for the window material for MEGAPIE.

* Corresponding author. Address: Paul Scherrer Institut, CH-5232 Villigen PSI, Switzerland. Tel.: +41-56 310 4712; fax: +41-56 310 2199.
E-mail address: heike.glasbrenner@psi.ch (H. Glasbrenner).

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Hence, the behaviour of T91 material in the presence of static load, irradiation and flowing Pb–Bi up to 300 °C has to be investigated before a liquid metal target can be irradiated in a proton beam for an extended period of time. This investigation will be performed in the LiSoR loop [3,4] at PSI in 2002 and 2003. During the experiment, the specimen will be irradiated in a proton beam with 72 MeV under static stress in the presence of flowing Pb–Bi at 300 °C.

The phenomenon known as liquid meal embrittlement (LME) describes the ductility loss of stressed metals or alloys in contact with liquid metals and can lead to a severe and very fast usually brittle intergranular failure of the structural material. Recently, it was observed that lead can induce severe LME of the ferritic/martensitic steel T91 under specific conditions in liquid lead [5,6].

The effect of pure bismuth on the mechanical properties of steels has not been widely studied. Tensile tests on mild steel in bismuth at 300 °C described by Rädecker [7] revealed no evidence of embrittlement but Tanaka and Funkunaga [8] observed significant decreases in the reduction in area of mild steel stressed at 350 and 460 °C and a recovery temperature, T_R of 550 °C.

Liquid metal alloys can cause embrittlement in the same way as pure metal. Additions of antimony, bismuth, copper or zinc can increase the embrittling potency of lead [7]. Legris et al. [5] reported that a notched and hardened T91 specimen showed a brittle fracture face after testing in Pb–Bi at 260 °C due to LME. Another martensitic steel CrNiMoVNb named MANET (Martensit for Net European Torus), foreseen as structural material for the first wall and blanket in a future fusion reactor, was intensively investigated with regard to future application [9–12]. Corrosion [13,14] and mechanical tests [15,16] on this steel were also performed in the presence of Pb–17Li. There seems to occur no LME for the combination MANET and Pb–17Li in the temperature range of 250 and 300 °C. Hence, the investigation of the mechanical behaviour of this material in Pb–55.5Bi environment is of great interest. Off-beam tensile tests were performed on MANET II steel in LBE and Ar during the commissioning phase of the LiSoR loop.

2. Experiments

2.1. Materials

The as received MANET II steel (produced by Saarstahl GmbH Völklingen, Germany; Heat No. 50806; composition listed in Table 1) was given the standard heat treatment (30 min at 1075 °C, air quenching and 2 h at 750 °C, air cooling). Composition is listed in Table 1.

Table 1
Composition of MANET II steel

Element	MANET II	Element	MANET II
Al	70 ppm	N	320 ppm
B	75 ppm	Nb	0.16
C	0.10	Ni	0.657
Co	0.005	P	0.004
Cr	10.37	S	0.005
Cu	0.01	Si	0.18
Mn	0.76	V	0.21
Mo	0.58	Zr	0.053

Impag AG (Switzerland) supplied the eutectic Pb–55.5Bi (44.8 wt% Pb and 55.2 wt% Bi) alloy which contained a few ppm of impurities: Ag 11.4, Fe 0.78, Ni 0.42, Sn 13.3, Cd 2.89, Al 0.3, Cu 9.8, Zn 0.2.

2.2. Test conditions

A series of flat standard tensile specimens were prepared with the following dimensions: thickness 1 mm, gauge length 20 mm, width 5 and 35 mm distance between shoulders. The specimens were degreased with ethanol before assembled into the circulating Pb–Bi loop LiSoR. The specimens were then exposed about 2 h at the particular testing temperature i.e. 180, 200, 250 and 300 °C before mechanical testing. The flow velocity of the eutectic melt was around 1 m/s in the test section and the flow conditions were turbulent. For comparison of the tensile test results achieved in Pb–Bi, tests were also carried out in the same facility but in an Ar atmosphere at RT, 180, 200 and 250 °C. The loop is not appropriate for operating at 300 °C with Ar, hence 250 °C was the highest temperature reached in the Ar environment. The tensile tests were performed at a constant cross head speed of 10^{-4} mm/s using a machine with a pneumatic jack that is regulated by an electromagnetic pressure valve. Some restrictions on the control of the tensile machine has limited the quality of these tests but a comparison can be done between the stress–strain curves achieved in Pb–Bi and Argon.

LiSoR experimental facility is an automatically operating LBE loop equipped with an electromagnetic pump, a flow meter, a heat exchanger system, an automatically fill and drain system and a load machine connected to the test section. More details of LiSoR loop are given in [3,4].

After tensile testing, the specimens were cut and prepared for analytical investigations by optical microscopy. SEM and EDX were performed on fracture surfaces as well as on the cross sections of the tensile probes. One sample was withheld to remove adherent Pb–Bi of the sample surface in order to see if Pb–Bi had penetrated into the structural material.

3. Results

3.1. Tensile tests

Fig. 1 reports the stress–strain curves obtained for the MANET II specimens tested in Ar at various temperatures. As temperature is increased, there is a decrease in ductility and yield strength for MANET II. The change between 180 and 250 °C is not so pronounced due to the fact that the temperature increase is slight.

The stress–strain curves after exposure to LBE show a dependence on temperature as well (Fig. 2). The difference between 180, 200 and 250 °C is visible but not well pronounced. A comparison of the stress–strain curves obtained at 180 and 200 °C in LBE and Ar environment shows no influence of LBE on mechanical behaviour of MANET II. The curves are nearly congruent (see Figs. 3 and 4). On the other hand, the ductility of the specimens tested in Ar and LBE at 250 °C seems to be influenced by the testing environment. The stress–strain curves after exposure to LBE at 250 and 300 °C compared with the curves obtained in Ar at 250 °C are plotted in Fig. 5. The ductility of the steel was reduced due to the contact of the liquid eutectic melt.

3.2. Fractography

The SEM micrographs of the specimens tested in Ar at any temperature show spherical dimples characteristic of ductile fracture resulting from uniaxial tensile load (an example is presented in Fig. 6: specimen tested at 200 °C in Ar).

In contrast, the fracture type observed on the specimens that failed in LBE is dependent on the testing temperature. The specimens tested at 180 and 200 °C (see Fig. 7) show the typical dimple structure, i.e. a ductile failure has occurred. This is in agreement with the measured stress–strain curves.

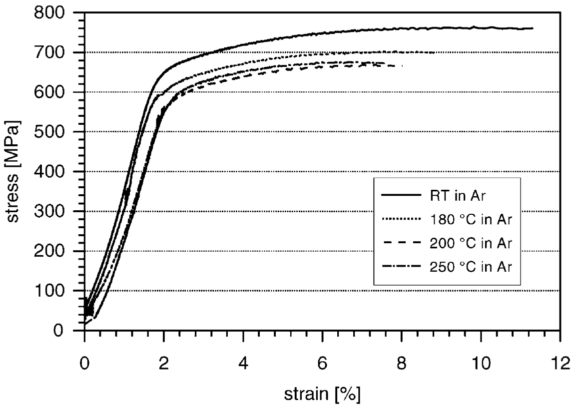

Fig. 1. Tensile test results of MANET II specimens failed in Ar at different temperatures.

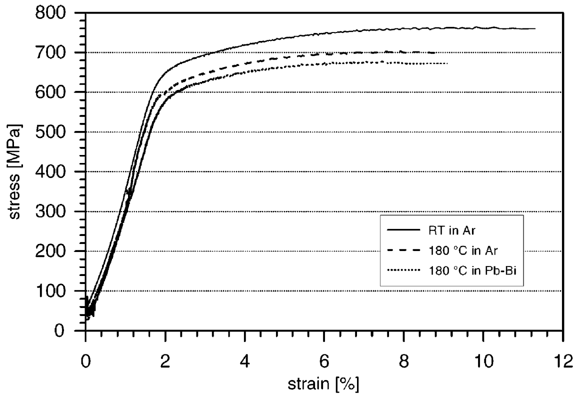

Fig. 3. Comparison of stress–strain curves obtained in Ar and LBE at 180 °C.

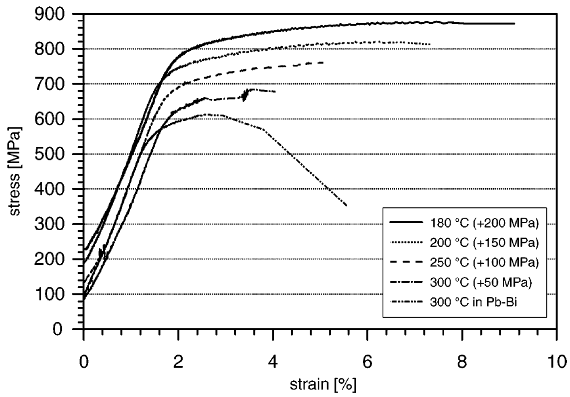

Fig. 2. Stress–strain curves obtained on MANET II at different temperatures in LBE.

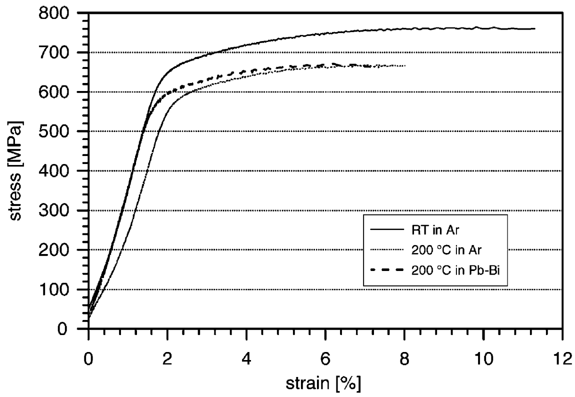

Fig. 4. Tensile test results of failed MANET II steel at 200 °C in Ar and LBE environment.

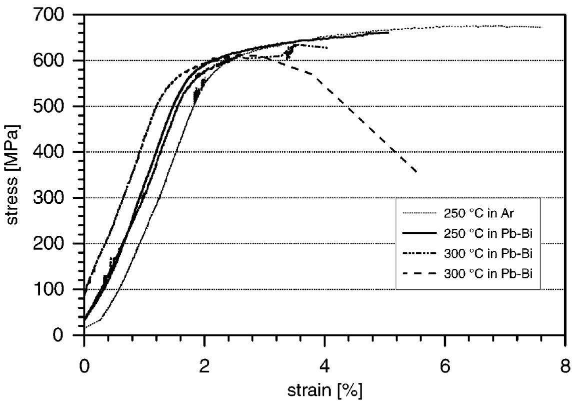

Fig. 5. Comparison of stress–strain curves revealed in Ar and LBE at 250 and 300 °C.

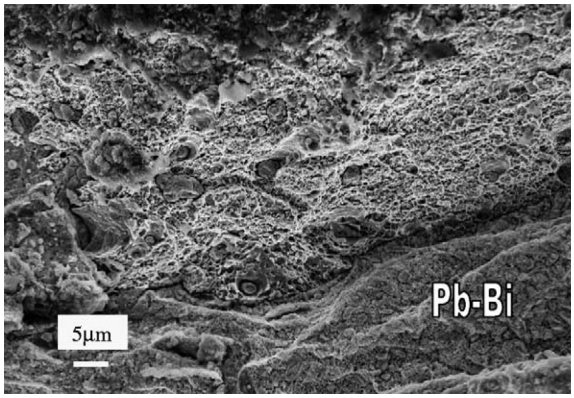

Fig. 8. Fracture surface of the MANET II specimen tested at 250 °C is quite well covered with solidified LBE.

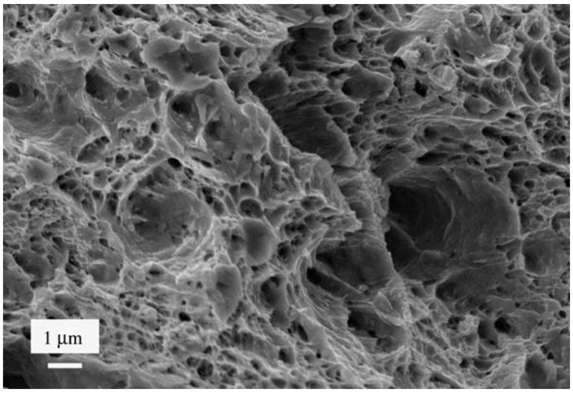

Fig. 6. Dimpled fracture surface of MANET II torn up in Ar at 200 °C.

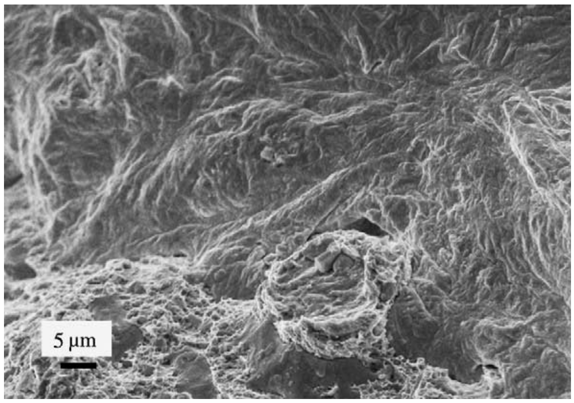

Fig. 9. SEM image showing the mixed fracture face of a MANET II specimen failed at 300 °C in Pb–Bi.

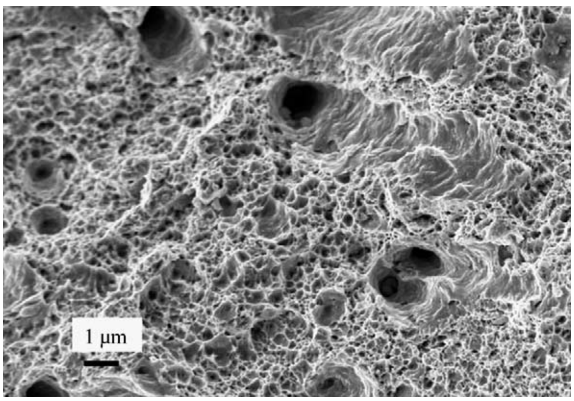

Fig. 7. Ductile fracture of MANET II tested in LBE at 200 °C.

The examined fracture surfaces of the specimens tested in LBE at 250 and 300 °C show a mixed fracture surface. The MANET II that failed at 250 °C has a ductility loss at the periphery but in the centre the visible fracture face is ductile. Adherent LBE was detected on some areas of the fracture faces due to the fact that it was not removed after testing and the fracture face of the MANET II specimen tested at 250 °C in LBE was especially well covered by solidified LBE (Fig. 8). On the specimens tested at 300 °C ductile and brittle areas are revealed all over the fracture surface, but there is no pattern concerning the location of brittle or ductile areas (Fig. 9). The brittle fracture is flat with no dimples and corresponds to a cleavage-like failure, however, the ductile rupture face shows the typical dimple structure as can be found on the Ar specimens.

3.3. Cross section examination

MANET II tensile specimens tested in LBE and Ar were longitudinally polished for metallurgical examination and SEM/EDX analysis. Again adherent LBE was

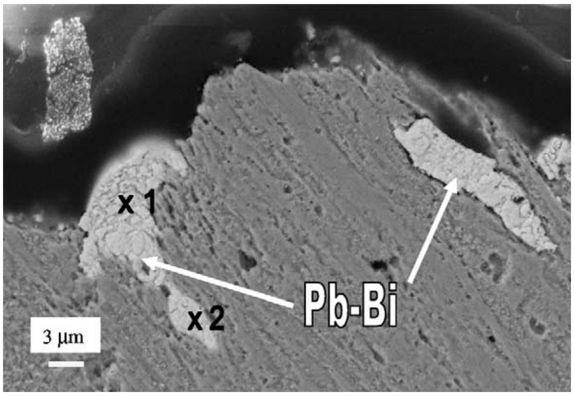

Fig. 10. SEM observation of on the cross section of specimen tested at 250 °C revealed penetration of LBE into the structural material.

not removed. Optical inspection of the etched cross sections reveals a martensitic structure for all specimens. Solidified Pb–Bi is observed on some areas of the fracture surface of the specimens tested in LBE at 180 and 200 °C, but no penetration of the LBE into the structural material is visible. On the other hand the cross section of specimens failed at 250 and 300 °C in LBE indicate penetration of Pb–Bi into the base material. A SEM image is given in Fig. 10. An EDX analysis of the penetrated alloy reveals that an enrichment of Bi has occurred compared to the eutectic composition Pb–55.5Bi. In point 1 (see Fig. 10) only the elements Pb and Bi can be recognised, in point 2 the steel elements Fe and Cr were detected additionally.

4. Discussion and conclusions

Mechanical tensile testing clearly shows that the ultimate tensile strength of MANET II steel decreases with increasing temperature. This is the expected behaviour for martensitic steels. Additionally, the environment can intensify this effect due to LME.

The fracture surfaces of the MANET II specimens tested in Ar are dimple structured indicating fully ductile fracture. The specimens tested at 180 and 200 °C in LBE show as well a ductile fracture. The congruence of the stress–strain curves revealed at 180 and 200 °C in LBE and Ar confirms the ductile fractures as well.

Pb–Bi eutectic melt embrittles MANET II steel at least at 250 and 300 °C. The fracture surfaces of the specimens tested in liquid LBE remained partly covered by solidified melt. Nevertheless, a mixed fracture mode with brittle and ductile regions was observed on the fracture surfaces of the specimens tested. Also the stress–strain curves confirm the influence of LBE on the mechanical properties.

Hence our observations are in disagreement with the common statement that LME normally is most pronounced around the melting point of the embrittler, i.e. in our case $T_m = 125$ °C, the melting point of Pb–Bi.

The results presented by Balandin et al. [17] are also contrary to this general statement that LME is most severe around the melting point of the liquid metal. They found the largest reduction in the ductility of pearlitic type 12KhM, a Russian steel, tested in oxygen-bearing LBE at around 400 °C. At 200 °C the mechanical properties of this steel were independent of the testing environment LBE or Ar. The embrittling effect of the liquid Pb–Bi disappears at 600 °C. There is no oxygen control or detection system in LiSoR. But we can assume that the oxygen content in LBE is close to saturation, because no special effort was made to reduce the oxygen content in the system. Hence the comparison of our results and test conditions with those of Balandin shows excellent agreement: no LME was found at 180 and 200 °C; Pb–Bi embrittles MANET II at 250 and 300 °C.

LME is not unexpected for the combination MANET and Pb alloy. Although tensile tests performed on fully tempered MANET I steel in static Pb–17Li showed no LME at 250 and 300 °C [15,16], the reason can be the use of a somewhat different liquid metal alloy. Bismuth is a more aggressive corrodent than lead. In general, metals are more soluble in bismuth. We analysed the penetrated melt in the steel matrix, and it was enriched in Bi compared to the eutectic composition Pb–55.5Bi. This observation is in total agreement with the results presented from Fazio et al. [18] who has found as well an enrichment of Bi in the penetrated LBE melt in a T91 steel matrix. So it seems that the element Bi is responsible for the embrittling of the steel. A SIMS analysis will be conducted on the alloy penetrated into the steel in order to quantify the Pb–Bi ratio.

Another parameter influences the appearance of LME: the extent of wetting of the specimen surface by the liquid Pb–Bi alloy. It has been established that LBE embrittles steel only when the surface can be wetted by molten alloy because the embrittler must be transported to the tip of the propagation crack, i.e. embrittlement occurs only when the crack tip is wetted by the liquid metal. Adsorption of Pb–Bi atoms to the grain boundaries reduces the cohesive strength of the iron–iron bonds in the boundary and as a consequence a lower stress concentration is required for crack initiation. It is well-known that LME is, among other things, dependent on the time and temperature of exposure to achieve good wetting. Before rupture testing, the MANET II specimens were not pre-wetted and the exposure to LBE was only around 2 h at the test temperature. Hence, it could be argued that LME was not observed at 180 to around 200 °C because the steel specimens were not wetted by LBE and hence Pb–Bi could not penetrate along the grain boundaries. Wetting is dependent on time and temperature; this could mean that the tensile specimens showing LME were already wetted at 250 and 300 °C after 2 h of exposure. Nevertheless this has to be checked because it seems unlikely that the native oxide of the MANET surface will be dissolved so quickly.

Anyway, future tensile tests should be performed with pre-wetted MANET II. The influence of the strain-rates and impurities in LBE (mainly oxygen) on the mechanical behaviour of the steel is of great interest and should be investigated as well.

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