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TWIN ASTIR: First tensile results of T91 and 316L steel after neutron irradiation in contact with liquid lead–bismuth eutectic

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A B S T R A C T

The TWIN ASTIR irradiation program [1] was aimed at determining the separate and possibly synergistic effects of a liquid lead–bismuth eutectic (LBE) environment and neutron irradiation. The materials in one capsule were irradiated in contact with LBE to a dose of about 1.5 dpa at a temperature between 460 and 490 °C and subsequently tested in liquid lead–bismuth eutectic environment at temperatures between 200 °C and 450 °C. This paper discusses the tensile results of T91 and 316L with and without irradiation while in contact with liquid lead–bismuth eutectic. The ferritic–martensitic T91 was softened by 50–100 MPa while the austenitic 316L steel showed a slight decrease in total elongation. Although no irradiation hardening was observed at the elevated irradiation temperature, the ferritic–martensitic T91 steel was prone to a certain extent of liquid metal embrittlement at 200 °C, 350 °C and 450 °C which was not affected by irradiation. The decrease in total elongation of the 316L is thought to be due to the neutron irradiation effect.

1. Introduction

Lead–bismuth eutectic was selected to be both coolant and spallation target material for MEGAPIE [2] and for several future experimental accelerator driven systems (ADS) around the world (MYRRHA or XT-ADS in Europe [3,4]; J-PARC in Japan [5], ADS in India [6] and HYPER in South Korea [7]). This new concept of reactor type might be one of the possible solutions for the nuclear waste problem as it is conceived to be able to transmute high level radioactive waste and long lived actinides [8].

The ADS technology however requires special operating conditions. The materials need to withstand temperatures ranging between 200° and 550 °C under a high energy neutron flux and in contact with the liquid lead–bismuth eutectic (LBE). This liquid metal contact does not only result in liquid metal corrosion but might also facilitate liquid metal embrittlement (LME).

While both the LME phenomenon and liquid metal corrosion in lead–bismuth eutectic are currently widely under investigation by numerous laboratories around the globe, little is known about the possible synergy between irradiation and liquid metal corrosion and embrittlement. There is a lot of experience in the field of liquid sodium from the fast reactor community; however the current knowledge on liquid lead–bismuth technology is still rather scarce and mostly limited to the military experience of the Russians.

Materials are known to undergo irradiation hardening when subjected to neutron irradiation below their Stage V-temperature which may make them more prone to liquid metal embrittlement as it was observed by Vogt et al. when the ferritic–martensitic T91 material was hardened by a dedicated heat treatment [9].

Recently however, Sapundjiev et al. [10] performed slow strain rate tensile tests (SSRT) in LBE and in air on tensile specimens that were irradiated in contact with water to about 1.5 dpa at 200 °C. Both the tests and the neutron irradiation of the examined T91 and 316L steels were performed at 200 °C. The SSRT tests were performed using a strain rate of $5 \cdot 10^{-6}$ s^{-1}. No effects of the LBE environment were found in this work as compared to the results on the same materials tested in air. The authors did remark that the irradiation hardening effect in the studied material ($\Delta\sigma_Y$ 316L: 155 MPa; $\Delta\sigma_Y$ T91: 260 MPa) was far less than observed in the studies performed on thermally hardened T91 ($\Delta\sigma_Y > 500$ MPa for HT500) [11,12] which did show LBE embrittlement. Furthermore, the samples used in the LME study after neutron irradiation by Sapundjiev et al. did not have any surface stress concentrators in contrast to the EDM cut samples used by Dai and co-workers [11,12], that showed substantial susceptibility to LME. Apart from the hardening effect and the absence of surface stress concentrators, one needs to keep in mind that these samples were irradiated in water and subsequently tested in LBE without long term exposure to low oxygen containing LBE prior to the actual SSRT test.

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Table 1
Chemical composition of the tested T91 and 316L steels, wt.%.

Material	B	C	N	Si	P	S	V	Cr	Mn	Ni	Nb	Mo
T91	<1 wppm	0.1	0.04	0.22	0.021	0.0004	0.21	8.99	0.38	0.11	0.06	0.89
316L	<1 wppm	0.019	0.03	0.67	0.032	0.0035	0.07	16.7	1.81	9.97	–	2.05

Thus it is probable that the oxide layer which was built up during irradiation in water would have been thicker than the native oxide layer present on the non-irradiated sample and might also have had an important role in prevention of liquid metal wetting in these tests. In fact, it was shown previously that absence of wetting due to the presence of the native oxide layer on ferritic–martensitic T91 steel prevents the occurrence of LME in liquid lead–bismuth eutectic environment [13]. Therefore, it is more than likely that the oxide layer on the samples irradiated in water up to 1.5 dpa prevented the occurrence of LME. Unfortunately, no examinations were made to characterize the oxide layer on the sample before and after testing in LBE.

The irradiation experiment called TWIN ASTIR, which stands for ADS Steel T91 Irradiation up to two doses (hence Twin) was the first neutron irradiation experiment in Europe where capsules filled with structural materials in contact with liquid LBE were placed inside a critical reactor. The experiment started receiving neutron irradiation during the second cycle, 2006 of BR2 in April 2006 [1].

In this paper, we will discuss the first tensile results of the ferritic–martensitic steel T91 and the austenitic steel 316L, irradiated in contact with LBE in capsule A of the TWIN ASTIR irradiation experiment and tested in contact with liquid LBE at temperatures between 200 and 450 °C.

2. Experimental

The chemical composition of both the ferritic–martensitic T91 steel and the austenitic 316L steel are given in Table 1. The original 15 mm thick 316L plate was solution annealed at 1050–1100 °C, followed by a water quench. The heat treatment of the original 15 mm thick T91 plate consisted of a normalisation treatment at 1100 °C for 15 min followed by a water quench to room temperature. The T91 plate was then tempered by heating the normalized steel to 770 °C for 45 min followed by air cooling to room temperature. These materials were irradiated and tested by using small scale cylindrical tensile specimens with a total length of 24 mm, a gauge length of 12 mm and a diameter of 2.4 mm. The tensile specimens were manufactured in L orientation (rolling direction) using a lathe machine and the gauge section of the specimens was polished to a mirror shine (up to P#4000 grid) prior to irradiation in contact with LBE. The environmental conditions and controls of irradiation experiment TWIN ASTIR were fully described in [1]. However, the actual irradiation temperature of capsule A was higher than previously reported [1] due to an underestimation of the gamma heating caused by the presence of LBE in the capsule. During the irradiation experiment, the temperature of the PWR water surrounding the capsule was monitored online. No temperature measurements were made inside the capsule during irradiation. The actual gamma heating values for capsule A of TWIN ASTIR are summarized in Table 2. This detailed calculation led to an irradiation temperature of the samples between 460 and 490 °C instead of 350 °C (327–368 °C) as mentioned in [1].

Moreover, since the gamma heating was significantly different in the structural materials compared to in the liquid LBE and the tensile samples were screwed into a retainer plate on one side only, the temperature varied over the sample. This is depicted in Fig. 1 which gives the temperature profile along the length of a sample in capsule A under irradiation. It should be noted however

Table 2
Actual gamma heating values for capsule A of TWIN ASTIR (in W/gr).

T91 sample	Inner SS tube	Outer SS tube	LBE
5.59	5.60	5.64	11.77

that the gamma heating values given in Table 2 are calculated for the middle of the gauge length and cannot be strictly applied to the entire geometry. The temperature profile across the sample would in actuality have been slightly more evened out compared to the profile presented in Fig. 1. The radial temperature distribution in the middle of the gauge of the tensile specimens for a single stage of three tensile samples screwed into their retaining plate is depicted in Fig. 2. These calculated temperatures are considered to have been the actual temperature distribution under irradiation and are confirmed by the first tensile results presented below.

Between irradiation cycles, the capsule was first cooled down to about 250 °C for about a day and subsequently slowly cooled down further to room temperature. Before starting a new cycle, the capsule was heated to 250 °C by the surrounding PWR water for about a day before receiving any neutron flux. The capsule returned to irradiation temperature in a matter of minutes while under irradiation.

The lead–bismuth eutectic filled capsule A of TWIN ASTIR was retrieved from the BR2 reactor after 6 reactor cycles which resulted in an accumulated neutron irradiation damage of about 1.5 dpa. The capsule was then stored in the BR2 deep pool at room temperature to allow cooling of the capsule by natural decay. After 11 months, the capsule was transported and dismantled inside the hot cell and the tensile samples, irradiated in contact with liquid LBE, were tested in contact with liquid LBE under controlled oxygen conditions (around 10^{-7} wt.%) in Limets 2 [15] without prior cleaning. All tests (in LBE and gas environment) were performed in Limets 2.

The applied strain rates were $1 \times 10^{-3}\ s^{-1}$ and $1 \times 10^{-6}\ s^{-1}$ for the tests at 200 and 350 °C. These strain rates were chosen in order to be above and below the transition strain rate for LME as reported by Hamouche-Hadjem et al. [16]. At 450 °C, the applied strain rate was $5 \times 10^{-5}\ s^{-1}$, which was the lowest strain rate to allow completion of the test within one working day. The latter was required due to the safety regulations for high temperature testing inside the heavy liquid metal technology hot cell. The oxygen concentration of the LBE in the test autoclave was kept around 10^{-7} wt.% for all applied test conditions of strain rate and temperature.

3. Results and discussion

3.1. Ferritic–martensitic T91 steel

Tensile tests were performed in liquid LBE environment at 200, 350 and 450 °C on T91 irradiated in contact with LBE between 460 and 490 °C to an accumulated damage of about 1.5 dpa. The test temperatures were chosen to cover the ductility trough of unirradiated T91 reported in [14]. Although certain contradictions are reported in the embrittlement effect of LBE on normalized and tempered, non-irradiated T91 at 200 °C (no embrittlement in [11,13] vs. embrittlement in [17]), most studies report embrittlement at 350 °C [11–13] with recovery of ductility between 450 and 500 °C [11,14]. Furthermore, the irradiation in contact with low oxygen

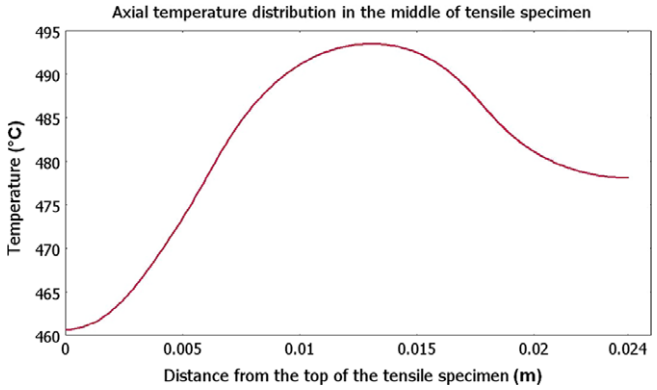

Fig. 1. Axial temperature distribution in the middle of a tensile specimen under irradiation in contact with liquid LBE inside capsule A of TWIN ASTIR (distance in meters).

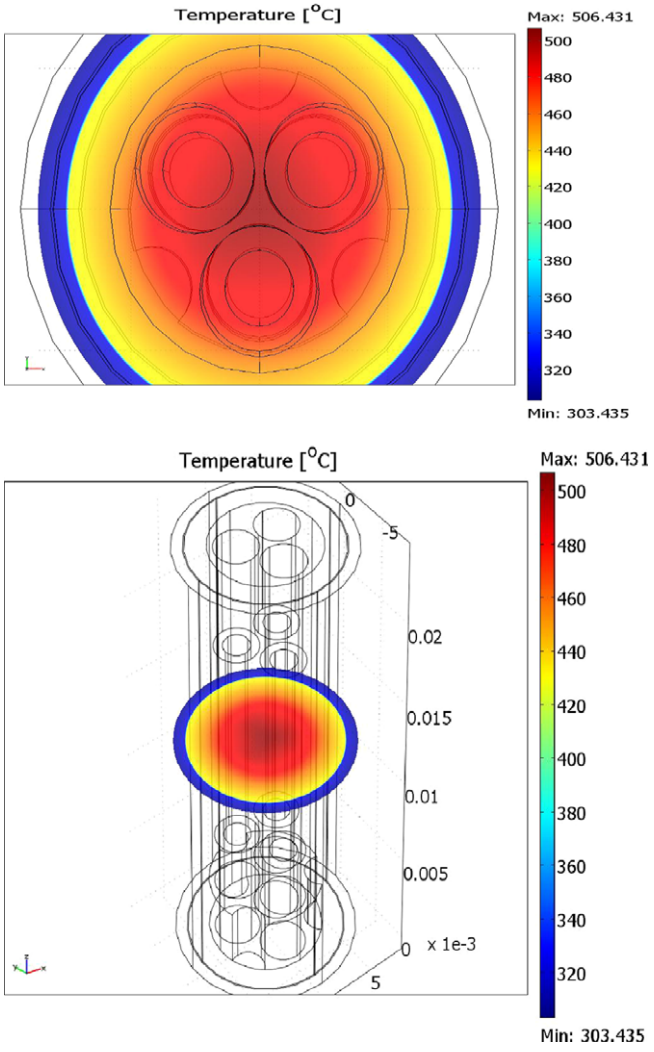

Fig. 2. Radial temperature distribution under irradiation in contact with LBE in the middle of the gauge of a stage of three tensile samples screwed into their retaining plate.

containing LBE was assumed to benefit wetting and therefore also increase the material's susceptibility to LME. The obtained tensile results of T91 steel after neutron irradiation at 460–490 °C while in contact with liquid LBE are depicted in Fig. 3. For the tensile tests performed at 200 °C (upper left in Fig. 3) there is no difference in yield strength or tensile strength between the reference test performed in a gas mixture of argon and 5% hydrogen and the test performed in liquid LBE environment after irradiation in liquid LBE. The total elongation is however reduced by about 5% for the test in LBE after irradiation at 460–490 °C in contact with LBE. At 350 °C (see bottom graphs in Fig. 3) there is a clear difference between the tensile curves in liquid LBE after irradiation at 460–490 °C tested at different strain rates. All curves show a decrease in yield strength and tensile strength (50–100 MPa) but the tensile curves resulting from the tests in LBE at a strain rate of $1 \times 10^{-6}\ s^{-1}$ show a strong decrease in total elongation while the curve at a strain rate of $1 \times 10^{-3}\ s^{-1}$ does not. This is in agreement with the brittle to ductile transition behaviour of T91 in contact with liquid LBE observed by Hamouche-Hadjem et al. at 160 °C [16]. Although the statistical significance is still very limited, the similar result obtained from both tensile tests at 350 °C, using a strain rate of $1 \times 10^{-6}\ s^{-1}$, after irradiation in LBE at 460–490 °C indicates improved wetting due to the irradiation in contact with LBE. As seen in the results of the as received tests in contact with LBE at 350 °C, LME was not always observed in T91, even under LME favourable conditions, due most probably to an absence of wetting.

At 450 °C the tensile results after irradiation in contact with LBE show a slight decrease in yield strength and total elongation of around 30–50 MPa. The obtained total elongation is similar to that measured on the reference test of non-irradiated T91 tested in LBE. The decreased total elongation compared to the reference test in air is slightly larger than the expected experimental scatter under these conditions and is attributed to LME. Based on the observed decrease in total elongation at 200, 350 and 450 °C, which is similar to this observed in T91 tested in LBE prior to irradiation, no synergistic effects seem to be occurring between LME and neutron irradiation while in contact with LBE.

Rather limited data is available on neutron irradiation effects in T91 above 400 °C. While Klueh et al. [18,19] report a stable microstructure of T91 when irradiated to 10 and 23 dpa at 450, 500 and 550 °C which resulted in the absence of hardening or softening, Yamamoto et al. [20] does report softening up to 100 MPa for ferritic–martensitic steels irradiated above 450 °C.

The observed softening of the neutron irradiated T91 at 350 and 450 °C in this work is in agreement with the data shown by Yamamoto et al. [20] on ferritic–martensitic steels irradiated in gas or water environment for irradiation temperatures above 450 °C. Although Yamamoto et al. [20] only includes data were the test temperature is equal to the irradiation temperature, our data show that the softening is actually more pronounced at 350 °C while it is not found when testing at 200 °C.

T91 does not seem to suffer from embrittlement due to precipitation growth and/or nucleation of separate phases caused by neutron irradiation above the irradiation hardening temperature as some other ferritic–martensitic steels were observed to do after irradiation up to about 5 dpa at 500 °C ($M_{23}C_6$ coarsening in Sandvik HT9, χ-phase precipitation in EM12, Laves phase formation in F82H) [21,22]. Furthermore, the observed decrease in total elongation is comparable to this observed when testing in contact with liquid LBE before irradiation. It is therefore considered very likely that the observed decrease in total elongation is due to LME. Fracture surface examination would be needed to confirm our findings. However, due to licensing issues this is currently not possible.

3.2. Austenitic 316L steel

Tensile tests were performed on 316L in liquid LBE environment at 200, 350 and 450 °C after neutron irradiation in contact with LBE between 460 and 490 °C to an accumulated damage of about 1.5 dpa. The material was tested at a strain rate between $1 \times 10^{-3}\ s^{-1}$ and $1 \times 10^{-6}\ s^{-1}$. Despite a change in fracture surface

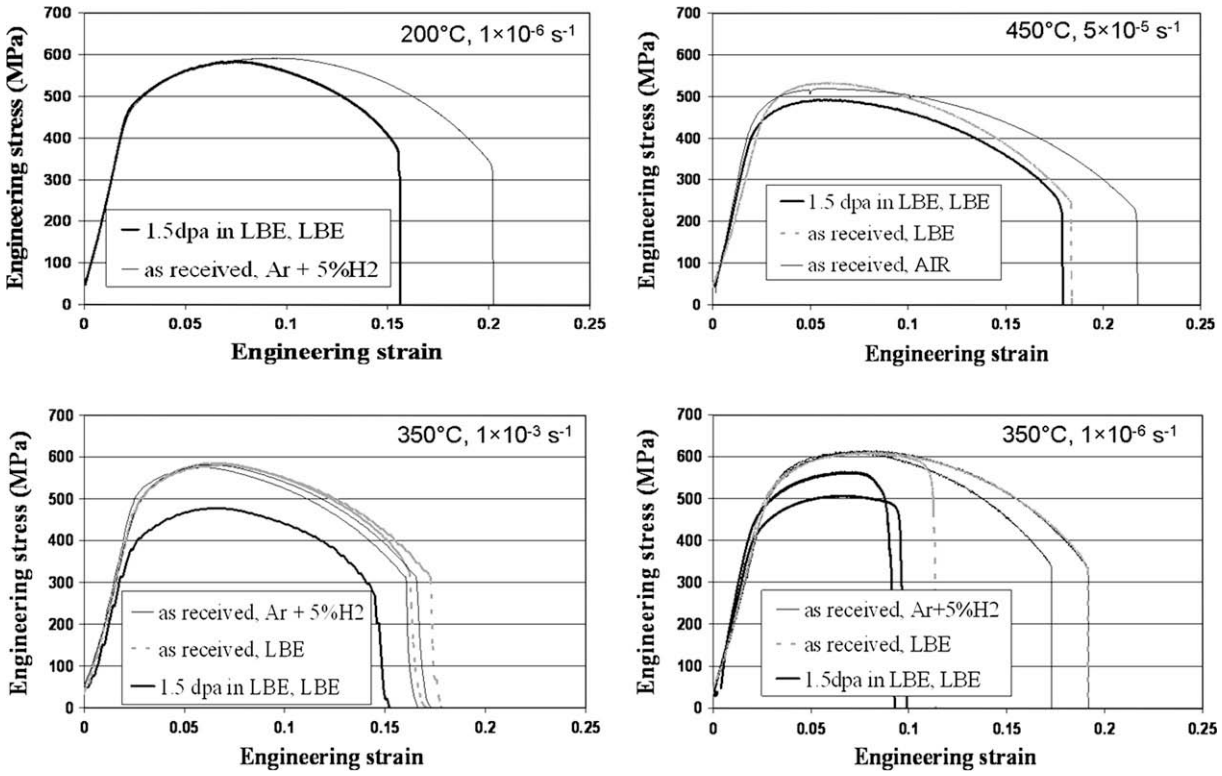

Fig. 3. Comparison of engineering stress–strain curves of T91 steel irradiated up to 1.5 dpa in contact with liquid LBE in capsule A of TWIN ASTIR and as received T91 steel (normalized and tempered), tested at 200, 350 and 450 °C in liquid LBE environment or gas environment using strain rates between 1×10^{-3} s^{-1} and 1×10^{-6} s^{-1}.

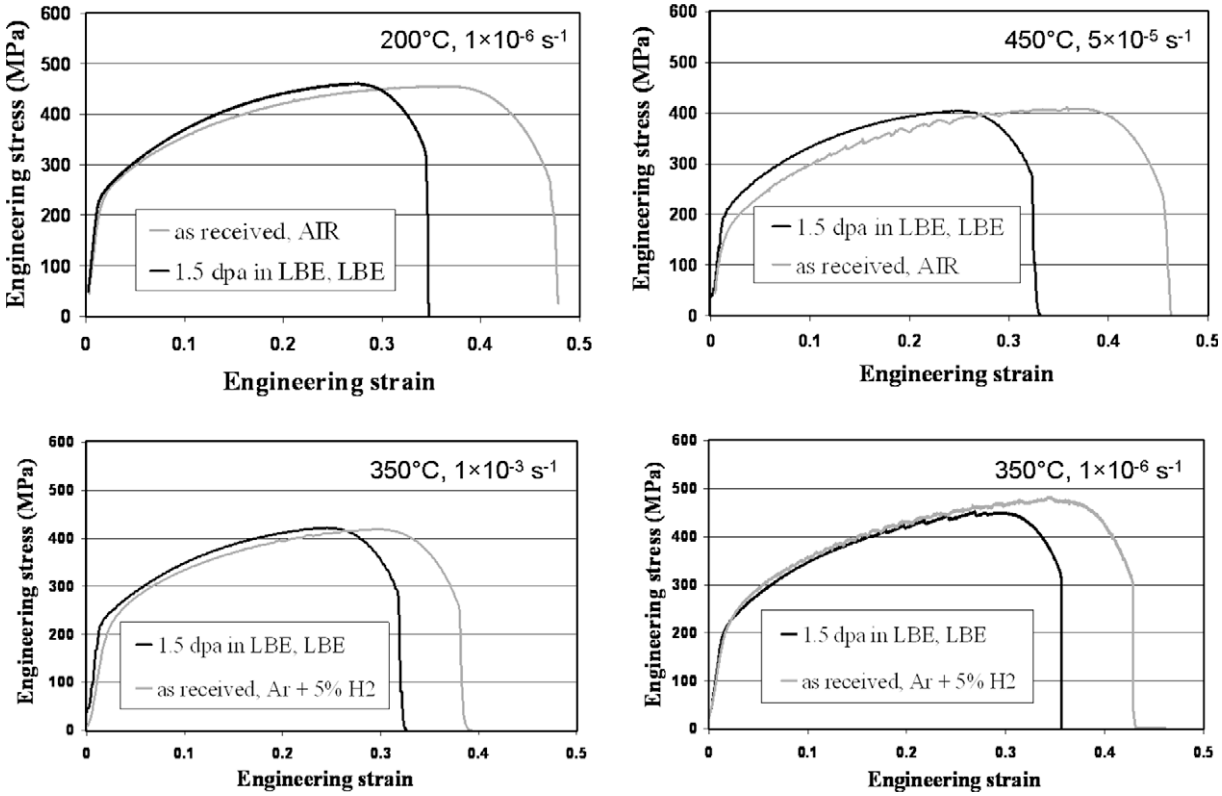

Fig. 4. Comparison of engineering stress–strain curves of 316L steel irradiated up to 1.5 dpa in contact with liquid LBE in capsule A of TWIN ASTIR and as received T91 steel (normalized and tempered), tested at 200, 350 and 450 °C in liquid LBE environment or gas environment using strain rates between 1×10^{-3} s^{-1} and 1×10^{-6} s^{-1}.

appearance observed in certain studies of 316L in contact with LBE [16], there is no reported change in mechanical behaviour of solution annealed, non-irradiated 316L when tested in contact with LBE. Therefore, testing conditions were chosen similar to those applied to T91. The comparison of the tensile curves of 316L obtained by testing in LBE after irradiation in contact with LBE at 460–490 °C to the reference tests of 316L tested in gas are depicted in Fig. 4.

Under all tested conditions of temperature and strain rate, the yield strength of the 316L steel remained largely unchanged. At 450 °C however, there is a slight increase in yield strength (about 30 MPa). Furthermore, the strain ageing effect seems to be removed after irradiation when testing at 450 °C. The most remarkable results are actually that for all test conditions the measurements of total elongation are significantly reduced while the slope and curvature up to the tensile strength remained nearly the same before and after irradiation at 460–490 °C in contact with liquid LBE. This type of mechanical behaviour is typically observed in LME of T91 in contact with LBE. However, due to the absence of LME in 316L before irradiation, LME is considered to be an unlikely cause of the observed reduction in total elongation.

Although the microstructure evolution of 316L under neutron irradiation is relatively well known for temperatures up to 370 °C due to the material's application in pressurized and boiling water reactors operating today, there is rather little data available on 316L for neutron irradiation temperatures above 400 °C. For 316 there is a significant amount of data available from the FFTF and EBR-II irradiations done in the US. However, in contrast to the slight increase in yield strength observed here when testing 316L at 450 °C after irradiation between 460 and 490 °C, the 316 irradiated in FFTF at 420 and 520 °C up to 44 dpa shows an increase in yield strength by more than 200 MPa when tested at the irradiation temperature [23].

A study by Maziasz [24] on the microstructure evolution of 316 under neutron irradiation at elevated temperature (400–700 °C) mentions the formation of eta phase (nickel and silicon rich diamond cubic phase) and Laves phase (Fe_2Mo) after irradiation to 54 dpa at 460 °C and the coarsening of $M_{23}C_6$ precipitates. On the other hand, Stanley et al. [25] mentions the formation of ferrite in the austenite matrix of 316 due to neutron irradiation at temperatures above 450 °C. All these changes possibly occurred in the microstructure of the 316L steel studied here. Since they would all have a certain influence on the mechanical behaviour of the 316L steel, it is very difficult at this point to determine the cause of the observed decrease in total elongation.

Unfortunately, strict regulations regarding the possible Po^{210} contamination of other facilities made it impossible up to now to examine the fracture surfaces or microstructure of the tested materials or to examine the corrosive interaction of the LBE with the solid structural steels after irradiation in contact with LBE. These examinations will be performed as soon as the necessary licensing procedures for handling polonium contaminated materials will be completed.

4. Conclusions

The first tensile results of the ferritic–martensitic steel T91 and the austenitic steel 316L, irradiated in contact with LBE at 460–490 °C in capsule A of the TWIN ASTIR irradiation experiment and tested in liquid LBE at temperatures between 200 and 450 °C were presented and discussed.

Although the increased irradiation temperature in capsule A was above the stage V-temperature of the examined steels and therefore did not cause any irradiation hardening, the irradiation in contact with low oxygen containing LBE did most probably improve wetting and therefore enable LME to occur.

Neutron irradiation in contact with LBE at 460–490 °C caused softening of the ferritic–martensitic steel T91 by 50–100 MPa when tested at 350 and 450 °C. No softening was observed at 200 °C. The observed LME of T91 by LBE after neutron irradiation in contact with low oxygen containing LBE was more reproducible after neutron irradiation, which might indicate improved wetting due to liquid metal corrosion. A certain extent of LME of T91 was observed at all tested temperatures while the degree of total elongation reduction was comparable with the reduction measured when tested in contact with LBE prior to irradiation. This confirms the absence of synergistic effects between neutron irradiation and LME under these conditions.

For the austenitic 316L steel, neutron irradiation in contact with LBE at 460–490 °C is not expected to have increased the material's susceptibility to LME. The observed decrease in total elongation after neutron irradiation in contact with LBE at 460–490 °C is thought to be due to the effect of neutron irradiation rather than due to the liquid metal contact.

Fracture surface examination by SEM will be performed in the near future to confirm these conclusions as soon as the necessary licensing procedures to examine polonium contaminated materials will be completed.

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References

[1] J. Van den Bosch, A. Almazouzi, Ph. Benoit, R.W. Bosch, W. Claes, B. Smolders, P. Schuurmans, H. A Abderrahim, J. Nucl. Mater. 377 (2008) 206–212.
[2] W. Wagner, F. Gröschel, K. Thomson, H. Heyck, J. Nucl. Mater. 377 (1) (2008) 12–16.
[3] L. Cinotti, B. Giraud, H. A Abderrahim, J. Nucl. Mater. 335 (2004) 148–155.
[4] H. A Abderrahim, P. Kupschus, E. Malambu, Ph. Benoit, K. Van Tichelen, B. Arien, F. Vermeersch, P. D'hondt, Y. Jongen, S. Ternier, D. Vandeplassche, Nucl. Instr. Meth. Phys. Res., A 463 (2001) 487–494.
[5] T. Sasa, Prog. Nucl. Energy 47 (1–4) (2005) 314–326.
[6] P. Satyamurthy, L.M. Gantayet, A.K. Ray, Pramana J. Phys. 68 (2) (2007) 343–353.
[7] Ch. Cho, N.-I. Tak, J.-H. Choi, Y.-B. Lee, Ann. Nucl. Energy 35 (7) (2008) 1256–1263.
[8] C. Rubbia, J.A. Rubio, S. Buono, F. Carmianti, N. Fietier, J. Galvez, C. Geles, Y. Kadi, R. Klapish, P. Mandrillioni, J.P. Revol, C. Roche, CERN Report AT/95-44 (ET), 1995.
[9] J.-B. Vogt, A. Verleene, I. Serre, A. Legris, J. Nucl. Mater. 335 (2004) 222–226.
[10] D. Sapundjiev, J. Nucl. Mater. 356 (2006) 229–236.
[11] B. Long, Z. Tong, F. Gröschel, Y. Dai, J. Nucl. Mater. 377 (1) (2008) 219–224.
[12] B. Long, Y. Dai, J. Nucl. Mater. 376 (3) (2008) 341–345.
[13] J. Van den Bosch, D. Sapundjiev, A. Almazouzi, J. Nucl. Mater. 356 (1–3) (2006) 237–246.
[14] Y. Dai, B. Long, F. Gröschel, J. Nucl. Mater. 356 (1–3) (2006) 222–228.
[15] R.W. Bosch, A. Almazouzi, D. Sapundjiev, H. Dekien, Development of a hot-cell test set-up for liquid metal embrittlement (LME) studies in lead–lithium and lead–bismuth, in: Proceedings of EUROCORR 2005, Lisbon, Portugal, pp. 1–8.
[16] Z. Hamouche-Hadjem, T. Auger, I. Guillot, D. Gorse, J. Nucl. Mater. 376 (2008) 317–321.
[17] J. Van den Bosch, G. Coen, A. Almazouzi, J. Degrieck, J. Nucl. Mater. 385 (2) (2009) 250–257.
[18] R.L. Klueh, J.M. Vitek, J. Nucl. Mater. 132 (1985) 27–31.
[19] R.L. Klueh, J.M. Vitek, J. Nucl. Mater. 182 (1991) 230–239.
[20] T. Yamamoto, G.R. Odette, H. Kishimoto, J.-W. Rensman, P. Miao, J. Nucl. Mater. 356 (2006) 27–49.
[21] R.L. Klueh, K. Shiba, M.A. Sokolov, J. Nucl. Mater. 377 (2008) 427–437.
[22] R.L. Klueh, K. Shiba, M.A. Sokolov, J. Nucl. Mater., 2009, doi:10.1016/j.jnucmat.2008.12.090.
[23] R.L. Klueh, J. Nucl. Mater. 233–237 (1996) 197–201.
[24] P.J. Maziasz, Scripta Metall. 13 (1979) 621–626.
[25] J.T. Stanley, L.E. Hendrickson, J. Nucl. Mater. 80 (1979) 69–78.