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ScienceDirect
Journal of Nuclear Materials 376 (2008) 307–311
journal of nuclear materials
www.elsevier.com/locate/jnucmat

Investigation of the sensitivity to EAC of steel T91 in contact with liquid LBE

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Abstract

The ferritic–martensitic steel T91 is one of the most promising material for application in the generation IV type reactors. However, there are critical issues, such as the susceptibility to damage of the steel in contact with the heavy liquid metals and their effect on the mechanical properties of structural materials. In this context, it was initiated a study of the boundary conditions, necessary to ascertain the sensitivity of the T91 to environmentally assisted cracking when loaded in contact with the liquid lead–bismuth eutectic. A series of tensile tests were carried out in a cell where the specimens were immersed in static LBE. Results showed that at high temperature the steel in contact with the liquid metal had a slight decrease of yield and UTS value and a marked increase in the elongation to rupture. However, at low temperature the elongation to rupture and the reduction of area decreased, indicating the sensitivity to EAC.

PACS: 61.25.Mv; 62.20.−x

1. Introduction

Environmentally assisted cracking, EAC, is a degradation phenomenon, which is characterised by the initiation and growth of cracks in materials under the simultaneous and synergistic interaction of mechanical stresses and chemically active environments. Although this phenomenon is widely referred to in cases of water chemistry, there are specific cases of interactions between structural steels and heavy liquid metals, HLM (such as Hg, Pb, Bi), where the phenomenon is named liquid metal embrittlement, LME.

In general, there are several variables affecting the sensitivity of materials to LME. In particular the temperature is an important variable, since the highest sensitivity to the environment is believed to be at about the melting point of the liquid metal [1,2].

Moreover, this phenomenon has been commonly associated with the failure of the oxide film on the surface of the steel and the direct contact between the liquid metal and the surface of the steel under stress. In fact, in laboratory testing, when the oxide is selectively removed from the surface of the steel and the liquid metal is in direct contact with the bare metal (wetting), it was observed a marked reduction of the ductility of the steel [3,4]. Moreover, factors such as hardening of the alloy or the presence of notches (stress concentrators) were also proved to induce a reduction of ductility when the steel was in contact with lead bismuth eutectic, LBE [5,6].

In terms of mechanism of LME, there are several theories that have been proposed for specific cases, such as specific couples metal/LM, temperatures, initiation and crack growth rates. There is a general agreement on the concept of an adsorption-driven phenomenon, because the high crack growth rates reported could not be justified by diffusion phenomena. This concept was introduced as the Rebinder effect, which postulates a reduction of the free energy caused by adsorption. The adsorption is described as the adhesion of molecules of media to the solid surface, resulting in relatively high concentrations of the molecules at the place of contact. Moreover, even if there is no definite evidence of the initiation of cracks in HLM, some of

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doi:10.1016/j.jnucmat.2008.02.029

the authors [7,8] referred rather to the induction time for the catastrophic LME to happen and agreed that in presence of a sufficiently deep, sharp crack a critical threshold value has to be reached before the fast propagation of the crack. The crack propagation induces the nucleation of dislocations and promotes intensive slip of them, which lead to the formation of voids for Lynch [8]; however, for other authors, the increased pileup of dislocations induces the localised hardening of the material, which ductility is reduced, as for the use of cold work [7].

At high temperature, outside the temperature range were LME could happen, Popovich and Dmukhovskaya [7] proposed that LME is inhibited because the effect of the adsorbed media is to increase the plastic flow, that is the ability of the piled up dislocations to cross grain boundaries and release the accumulated stress. However, based on Lynch hypothesis [8] the adsorption at the surface of the media, could induce an enhanced egress of dislocation and, thus, enhancement of plasticity.

The aim of this work was the screening of the effect of experimental variables on the mechanical properties of specimens immersed in LBE, under tensile load. In general, the contact with the LBE induced an increase in ductility in the ferritic steel T91 at high temperature; however, decreasing the testing temperature induced situations where the onset of cracks with cleavage-like fracture surfaces were locally observed. Although no marked differences were observed in the engineering values, initiation and propagation of cracks was affected by the environment, in the present experimental conditions.

2. Experimental

The composition of the martensitic steel T91 is listed in Table 1.

The plates of T91 were provided by Industeel, Arcelor group after undergoing normalisation and tempering heat treatments: normalisation at 1050 °C, 1 mm per minute, and then water-cooling. The tempering treatment was carried out at 770 °C for 45 min and then air-cooled. Tensile specimens were machined from the plates in the L direction. The round tensile specimens had a diameter of 4 mm and a gauge length of 20 mm, with a surface ground up to 600 grit finish.

The LBE had a composition of 44.5Pb55.5Bi [wt%].

The tensile tests were carried out in a closed vessel were the control of the environment was possible. Experiments were carried out in air and in the liquid LBE, with a flowing mixture of argon and 5% hydrogen. The temperatures used for these experiments, in both environments, were 300 °C and 500 °C. The vessel was installed in a 50 kN hydraulic loading machine. The tests were carried out at two different strain rates,

$$\dot{\varepsilon} = 10^{-4}\ \mathrm{s}^{-1} \text{ and } \dot{\varepsilon} = 10^{-6}\ \mathrm{s}^{-1}$$

The testing procedure was structured in a time scale such as the heating up period (up to the maximum temperature) was about 3 h; this was followed by about 15 h of stabilization, during which the system homogeneously reached the operating conditions, before carrying out the mechanical tests. Three specimens for each experimental condition were tested. The average values of the results are here reported.

After tests, the residual LBE deposits were removed from one half of the specimens and these specimens were used for examination of the fracture surface with optical and scanning electron microscopes. The other half of the specimen, with the residual deposit, was used for the study of the cross section. Measurements of reduction of areas, Z%, and total elongations to rupture, ε_{tot}%, were carried out and used as parameters to determine the sensitivity to EAC.

Table 1
Composition of the FM T91, wt%

Fe	C	Cr	Ni	Mo	Mn	Si	V
Bal.	0.1	8.9	0.1	0.9	0.4	0.2	0.2

3. Results

Several tensile tests were carried out for the same material T91, and specimen dimensions at different experimental conditions. After a general screening of the testing conditions (variation of temperatures, strain rates and environment) it was observed that the effect of the LBE on the mechanical properties of the material was a slight decrease of the engineering values, such as yield stress, σ_y, and ultimate tensile strength, σ_{UTS}, compared to the values measured in air (Table 2 summarises the main results of these tests).

However, the major difference was observed to be an increase in ductility for the specimens immersed in the LBE, at 500 °C, in terms of longer elongation to rupture (ε_{tot}%) compared to the tests in air. Nevertheless, the reduction of area (Z%) did not change markedly in both environments (Fig. 1).

Decreasing the testing temperature (from 500 °C to 300 °C), decreases in the ε_{tot}% and the Z% were observed when the specimens were immersed in LBE compared to air. In addition, by varying the strain rates, the ε_{tot}% and Z% decreased passing from high ($10^{-4}\ \mathrm{s}^{-1}$) to low strain rate ($10^{-6}\ \mathrm{s}^{-1}$). The most marked effect was observed in the values of the Z% at the low strain rate (Fig. 1).

The decrease in Z% and ε_{tot}% were directly related to the appearance of the fracture surface of the specimens (Figs. 2–4). In fact, for the specimens tested at a strain rate of $10^{-4}\ \mathrm{s}^{-1}$, at 500 °C (Fig. 2) and 300 °C (Fig. 3) there was a marked difference in the surface appearance. The specimens tested at 500 °C had the typical features of the ductile fracture; the cone-cup structure with the inner dimpled area and the outer shear fracture surface.

The cross section of several specimens showed that a thick oxide layer was formed on the surface of the steel.

Table 2
Summary of the values measured during the tensile tests

Number of specimens	T (°C)	Strain rate (s^{-1})	Media	σ_y (MPa)	σ_{uts} (MPa)	ε_{uts} %	ε_{tot}%	Z%
3	300	1×10^{-4}	Air	487	620	10.2	22.2	77
2	300	1×10^{-6}	Air	520	646	9.3	19.3	74
3	300	1×10^{-4}	LBE	484	614	9.8	20.3	70
3	300	1×10^{-6}	LBE	485	633	8	16.1	51
3	500	1×10^{-4}	Air	456	520	6.3	22.7	84
1	500	1×10^{-6}	Air	464	498	4.2	19.3	85
3	500	1×10^{-4}	LBE	418	489	5.4	30.5	85
3	500	1×10^{-6}	LBE	393	430	3.2	27.7	89

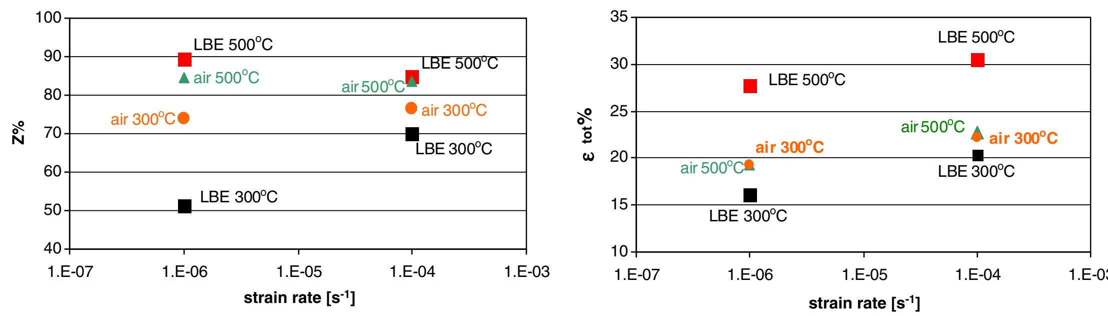

Fig. 1. Measurements of the reduction of area, Z%, and the elongation to rupture, ε_{tot} %, as a function of environment (air and LBE), temperature (300 °C and 500 °C) and strain rate (10^{-4} s^{-1} and 10^{-6} s^{-1}).

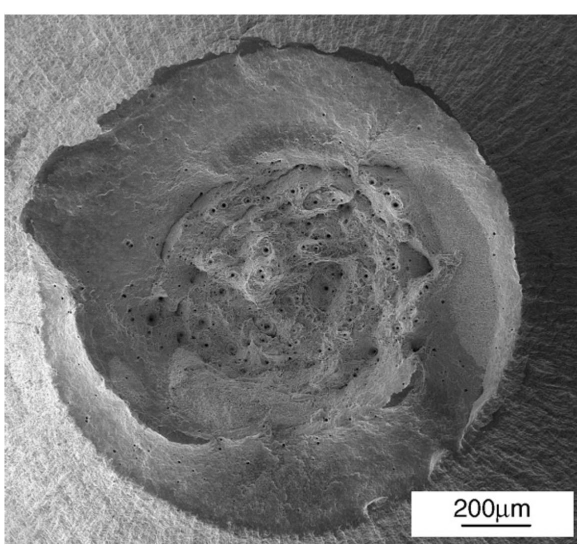

Fig. 2. Ductile fracture surface of specimens of the as-received ferritic steel T91 after rupture under tensile load in LBE with a strain rate of 10^{-4} s^{-1} at 500 °C.

The oxide was heavily damaged in proximity of the fracture surface and the liquid metal locally penetrated the metal. However, elemental analyses across these areas proved that an oxide layer was already formed at 500 °C and, thus, the intimate contact between the two metals was not reached.

On the other hand, the specimens tested at 300 °C had lateral cracks around the fracture surface, in the necked areas (Fig. 3(a)). The fracture surfaces of the lateral cracks had cleavage-like features (Fig. 3(b)), also visible in the whole fracture surface in several locations. However, the central part of the specimen failed in the ductile mode.

For the specimens tested at 300 °C and strain rate of 10^{-6} s^{-1} (Fig. 4), it was observed that one critical crack initiated the damage and had cleavage-like patterns. However, the remaining section of the specimen failed in the ductile mode.

4. Discussion

The aim of these tests was to ascertain the sensitivity of the martensitic 9Cr steel to EAC when in contact with LBE under tensile load. Results from the tensile tests were also complemented with the observation of the fracture surface of the specimens, in order to have a direct correlation between the mechanical properties measured and the characteristic features of the fracture surface.

Although the contact between the steel and the LBE was proved to induce changes in the fracture mode of the steel, in specific conditions (such as wetting, hardening and the presence of sharp notches) [1–4], in this work, these conditions were not imposed. For instance, the wetting between steel and LM was prevented because the oxygen present in the LBE was sufficient to develop a protective oxide layer on the steel, preventing their direct contact. However, by uniaxial loading of the specimens, the breakage of the surface oxide scale and the subsequent possibility of direct contact between the steel and the LBE were expected. This condition was not reached at higher temperature because of the oxygen contained in the liquid metal. At the highest

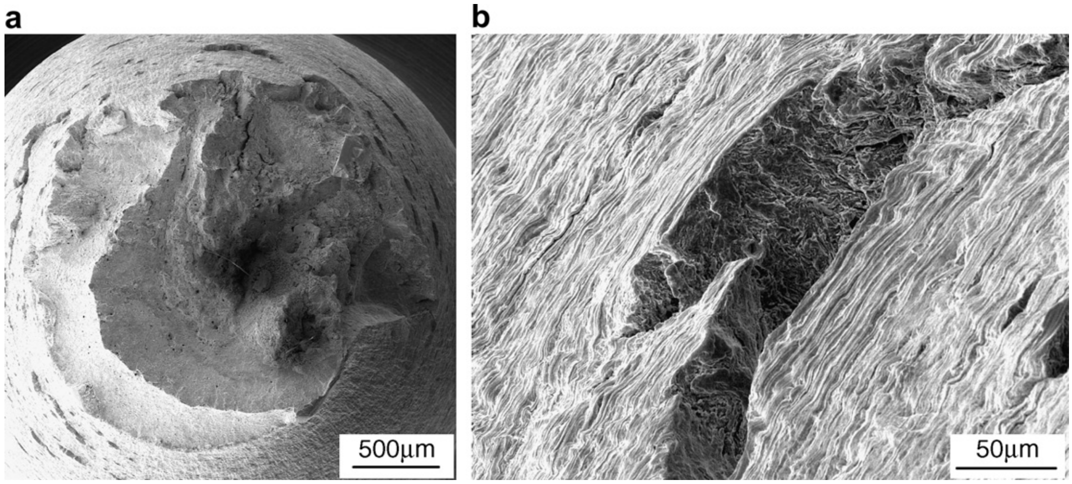

Fig. 3. (a) Fracture surface of specimens of the as-received ferritic steel T91 after rupture under tensile load in LBE at 300 °C with a strain rate of 10^{-4} s^{-1}; (b) detail of the initiation of the brittle fracture.

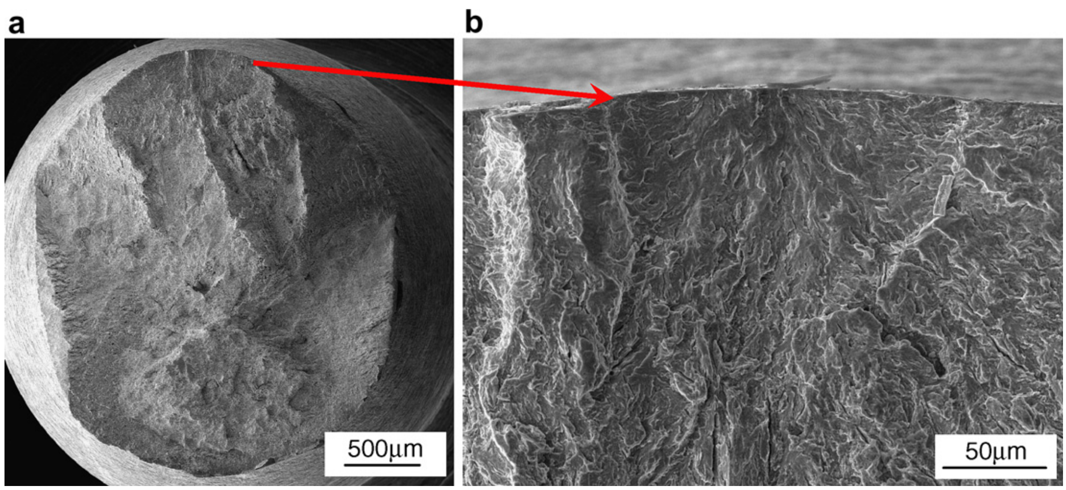

Fig. 4. (a) Fracture surface of specimens of the as-received ferritic steel T91 after rupture under tensile load in LBE at 300 °C with a strain rate of 10^{-6} s^{-1}; (b) detail of the initiation of the brittle fracture.

temperature (500 °C), the oxygen in the system was sufficient to rapidly self-heal the broken oxide on the steel and prevent the direct interaction between the metals. Other tests, carried out in similar conditions, in a reducing environment (flushing argon and hydrogen) [9] also showed the increased ductility at higher temperatures. These observations suggested that the interaction between the LBE and the steel at the oxidised surface was sufficient to modify the mechanical properties of the material, increasing its ductility. In fact, at the same temperature, but in air environment, the elongation to rupture was lower, excluding the effect of the temperature in terms of thermal activation.

On the other hand, by reducing the temperature and, thus, approaching the melting point of the eutectic, the behaviour of the material was markedly different and the initiation of cracks with brittle features was observed. In particular, at low strain rates the reduction of area was the lowest and the observation of the fracture surface confirmed that there were areas with cleavage-like features. Although at the surface of the specimen the conditions for the wetting were not possibly reached (high oxygen content in the media), there was evidence that localised areas were affected by the environment. This might suggest that the local conditions were changed/established during the test and was possibly related to the change in the local surface energy (possibly in correspondence to local inclusions or superficial defects). Moreover, the lower temperature might have reduced the kinetic of formation of the self-healing oxide creating a favourable condition for the direct contact between the metals once the initial oxide layer was damaged by the load applied.

This phenomenon was more evident for the specimen loaded at 300 °C and high strain rates (10^{-4} s^{-1}), where several lateral cracks with cleavage features were visible in proximity of the fracture surface, along the plastically deformed zone (necking of the tensile specimen).

In both experimental conditions, the initiation of cracks did not lead to a further increment of the cleavage-like rupture mode. In fact, even if the fracture facets appeared in the outer part of the specimen, the internal surface failed in the ductile mode, evidencing that the material was beyond the UTS point when it normally started necking and cracking. However, the cracks had brittle features whereas the plastically induced cracks have a totally different morphology.

Beside the environmental effect, the strain rate was observed to create different kind of response to the load. In fact, the specimens tested at high strain rates (fast tests) were more sensitive to crack initiation. However, at lower strain rates (slow tests) the crack increment was possibly higher. When one critical crack was initiated and the loading mode on the specimen changed the specimens were ruptured in shorter times (lower reduction of area and elongation to rupture).

5. Conclusions

In this study, the sensitivity to EAC of the steel T91 was observed when loaded in contact with LBE.

- The martensitic steel T91, when tested in tensile mode immersed in liquid LBE, was found to be sensitive to EAC.
- Sensitivity to EAC was observed at 300 °C, but not at 500 °C.
- At high strain rates, the steel was more sensitive to crack initiation, since a large number of brittle cracks were observed on the surface of the specimens.
- At low strain rates the number of lateral cracks was markedly reduced, but the elongation to rupture was shorter, suggesting a higher crack increment.

Acknowledgement

The authors are grateful to the European Union for funding the Project, 6.RP IP-EUROTRANS FI6W-CT-2004-516520, DM4-DEMETRA.

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