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Susceptibility to LME of 316L and T91 steels by LBE: Effect of strain rate

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Abstract

The effect of liquid lead–bismuth eutectic on 316L and T91 steels at 160 °C has been studied as a function of strain rate, using a centre cracked in tension specimen adapted for the study of crack propagation. Brittle fracture, characterized by elongated river cracks on all the fracture surfaces, indicates that T91 is sensitive to the embrittlement by LBE. This embrittlement effect is very pronounced at low deformation rate ($\sim 10^{-5}$ mm s^{-1}). A ductile–brittle transition is observed in the high strain rate range investigated. In the transitory regime, there is a competition between the growth of dimples and brittle cracking induced by the liquid metal. Ductility recovery is complete at the highest investigated deformation rate. The mechanical properties of the 316L steel are not clearly affected by the presence of LBE, in spite of a modification in the plastic deformation mode which strongly affects fracture surfaces.

1. Introduction

The T91 martensitic steel and the 316L austenitic steel are selected as candidates structural materials for an accelerator driven systems (ADS). The behaviour of these materials in the expected severe in-service conditions, namely neutron irradiation [1] and lead–bismuth eutectic (LBE) corrosion [2], is still under investigation. In addition to the corrosion issue, the phenomenon of liquid metal embrittlement (LME) is of particular interest as it may also impair the use of the selected material. LME is the premature brittle failure of a normally ductile material when it is strained in contact with liquid metal. The embrittlement manifests itself as a reduction in fracture stress, strain, or both [3]. In many cases, the liquid metal adsorption at the crack tip induces a ductile to brittle transition. In addition to plastic deformation, the most critical and mandatory condition for LME induction is that the liquid should be in intimate contact with the solid surface [4]. It should subsequently be present at the tip of the propagating crack to induce brittle fracture [5]. If the contact between the solid metal and the LM is hindered by the presence of an oxide film, LME is unlikely to occur. Due to their natural passive film, wetting of T91 and 316L steels by LBE is a crucial point in LME experiments and has hampered several studies of this phenomenon [6].

Few data exist on premature failure of 316L induced by liquid metal [7]. So far, only one study was undertaken on the 316L/LBE system [8] and there was little influence of LBE on the fatigue resistance of 316L. In contrast, the embrittlement by liquid Pb or LBE of ferritic/martensitic steels has already been reported [9,10]. In such case, unlike the classical LME example Al/Ga [11] or Cu/Bi [12], no inter-granular cracking was observed precluding high crack propagation rate and drastic reduction of strain to fracture in tensile tests.

LME depends on many parameters like the metallurgical state, the surface state, composition, solubility, temperature, strain rate... [4,13,14]. Temperature and strain rate are of extreme importance and will directly affect the severity of embrittlement of a given liquid metal/solid metal couple. It has been stated that LME appears in a certain range of temperature [4] and is more pronounced at higher strain rate [15]. One of the aims of this paper is to investigate the

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effect of strain rate on the embrittlement process of the two couples 316L/LBE and T91/LBE.

2. Experimental procedures

2.1. Materials and samples preparation

Two materials have been investigated: the austenitic stainless steel A316L and the modified 9Cr–1Mo martensitic steel T91. The steel compositions are reported in Table 1. The T91 steel was supplied by Creusot Loire Industrie with an austenitization treatment at 1050 °C followed by a tempering treatment at 750 °C. The 316L is a commercial austenitic stainless steel used in the annealed conditions. The lead–bismuth eutectic alloy (45 at.% Pb, 55 at.% Bi) used in this study was a 99.99% purity grade.

Centre cracked in tension (CCT) specimens, usually used for toughness testing, were machined as $150 \times 50 \times 1.5$ mm platelets of T91 and 316L. The centre elliptical notch of 10 mm in length and 2.5 mm width, was machined by electro-discharges-machining and then mechanically polished using SiC papers up to grade 4000 and electropolished. In order to obtain a good wetting, the oxide layer was dissolved using a soft soldering flux mainly composed of zinc chloride, ammonium chloride and hydrochloric acid, while the notch was filled with liquid LBE. This type of soft soldering flux is known to promote wetting of low melting point soldering alloys on steels [16,17].

In order to check that traces of flux do not affect mechanical properties of the T91 and 316L steels, mechanical tests on CCT specimens with the notch only filled by flux were also performed. In that case, the observed fracture mode remained always ductile.

2.2. Mechanical test

After the wetting pre-treatment, constant deformation rate tensile tests were used to characterize LME susceptibility. Tensile tests were performed, on an electromechanical testing machine MTS 20/MH, in an air-isolated test cell. A quartz tube was used to isolate the specimen in a stream of flowing He–4%H_2 (with less than 1 ppm oxygen). Specimens were heated by joule effect and were loaded at constant cross-head displacement rate, while the crack advance was recorded by a CCD TV camera. Loading was carried out at different cross-head displacement rates, going from 6.67×10^{-8} to 6.67×10^{-3} m s^{-1}. Reference mechanical tests were conducted using the same conditions, without liquid metal (LBE) in the central crack.

After a successful experiment has been carried out, examination of LME fracture surfaces shows complete coverage by the liquid metal. Liquid metal is in intimate contact with the solid and is usually difficult to remove. The solidified LBE was dissolved from fracture surfaces in a chemical mixture (1/3 ethanol, 1/3 acetic acid, 1/3 hydrogen peroxide); this process does not affect the fracture surfaces. Scanning electron microscopy (SEM) analyses of specimens and fracture surfaces were then performed on each sample after testing.

3. Results

3.1. T91 steel

3.1.1. Mechanical results

The mechanical behaviour is not affected by the presence of LBE, i.e. the yield and tensile strengths remain the same while the damage of T91 in contact with LBE is different from that observed without LBE, as can be see on Fig. 1(a). In a large range of deformation rate, T91 was found to be susceptible to LBE induced LME. In this geometry, strain localization occurs at both tips of the notch followed by the propagation of two symmetrical opposite macroscopic cracks until fracture occurs. In the presence of LBE, the cumulated plastic deformation before the crack initiation depends on the cross-head displacement rate (Fig. 1(b)). It is also observed that cracking and fracture occur more quickly, in the presence of liquid metal, at lower displacement rates. On the other hand, at high displacement rate, no difference was detected relative to reference tests (without LBE). These results strongly suggest the existence of a brittle to ductile transition controlled by the strain rate, which is confirmed by the fractographic analysis presented below.

3.1.2. Fractographic analysis

The fracture surfaces of specimens tested in contact with LBE, which illustrate the embrittlement effect, show multiple deep brittle cracks. Primary and secondary cracks, similar to elongated rivers, are formed and cover the entire crack propagation length (Fig. 2(a)). The insert of Fig. 2(a) reveals a multiple brittle crack initiation, in mixed mode I and II. The cracking mode is rather uncommon; it seems to proceed by shear bands decohesion, which is consistent with a plane stress shear deformation at 45° of the loading axis.

The proportion of the brittle fracture decreases with the increase of the cross-head displacement rate (Fig. 2(a)–(c)). In this transition regime, there is a competition between the

Table 1
Steels composition (wt%, balance Fe)

Elément	C	Ni	Cr	Mo	Cu	Si	S	Nb	Mn	P	V	Ti
T91	0.1	0.23	8.63	0.95	0.046	0.31	0.006	0.09	0.36	0.021	0.21	0.003
316L	0.022	10.20	16.70	2.10	–	0.48	0.003	–	1.32	0.025	0.06	–

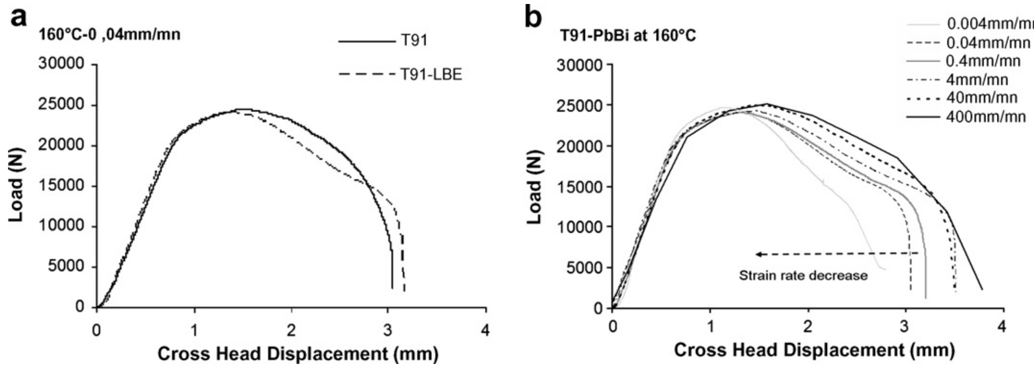

Fig. 1. (a) Load according to cross-head displacement obtained on T91 with and without LBE, 160 °C and 0.04 mm/min, (b) load cross-head displacement data as a function of deformation rate for T91 specimens with LBE, 160 °C.

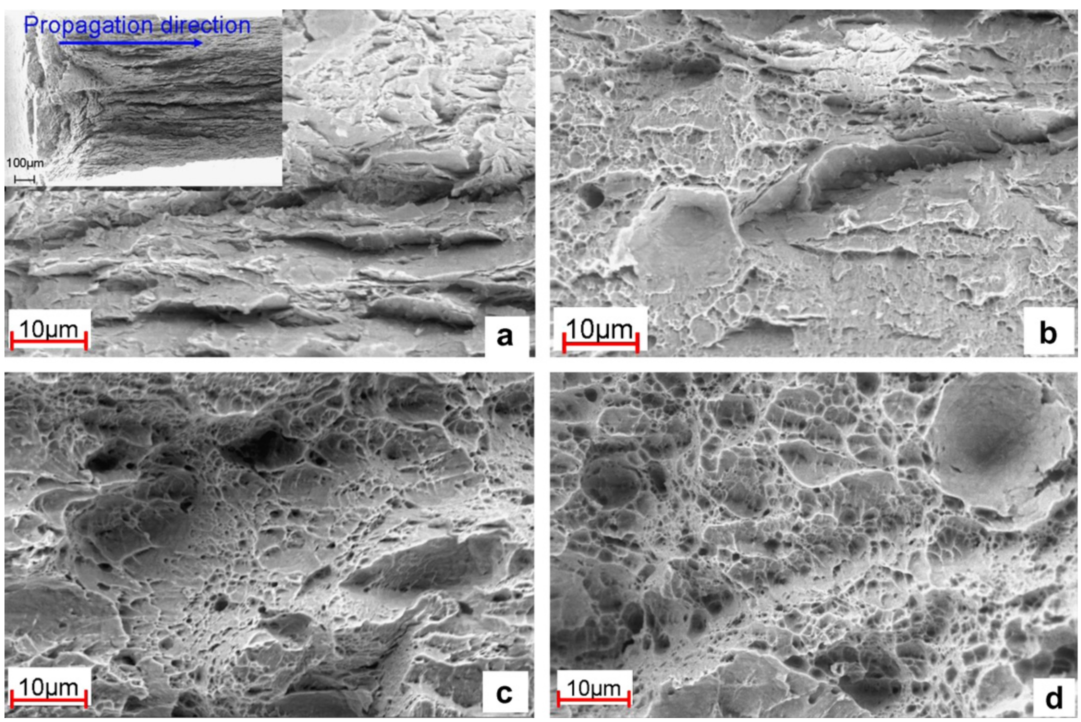

Fig. 2. SEM micrographs showing the fracture surfaces of T91 CCT specimens stressed at 160 °C (a) 6.67×10^{-7} m s^{-1} brittle regime in LBE, (b) 6.67×10^{-5} m s^{-1} brittle–ductile transition regime in LBE, (c) 6.67×10^{-3} m s^{-1} ductility recovery in LBE regime, (d) 6.67×10^{-7} m s^{-1} ductile fracture in He–4%H_2 (reference test). The crack initiation zone is reproduced in inset of (a).

growth of voids, the ductile rupture mode leading to dimples, and brittle cracking induced by the liquid metal. Ductility recovery is complete at the highest investigated displacement rate (Fig. 2(c)). The presence of LBE modifies the fracture mechanism of the T91 and inhibits the fracture mode by nucleation-coalescence of voids giving rise to the dimpled fracture surface observed in the absence of LBE, as seen in Fig. 2(d).

3.2. 316L steel

The fracture surface of the 316L tested in He–4%H_2 without LBE consists of small dimples typical of a ductile mode (Fig. 3(a)). In contrast, the fracture surfaces produced in contact with LBE are quite different, indicating a mixed brittle/ductile fracture profile: ductile by growth and coalescence of cavities and brittle by decohesion. These fracture surfaces are characterized by a ductile crack initiation stage (see the inset of Fig. 3(b)) and by the presence of primary and secondary cracks of various sizes connected by ductile zones (cavities) (Fig. 3(b) and (c)).

On the different SEM micrographs, the size and the number of cracks are different according to the deformation rate applied. It is noted that the embrittlement effect is more pronounced for the lowest rate.

The tensile curves were compared to ones obtained from testing 316L without LBE under the same conditions (Fig. 3(d)). There is no noticeable difference in the

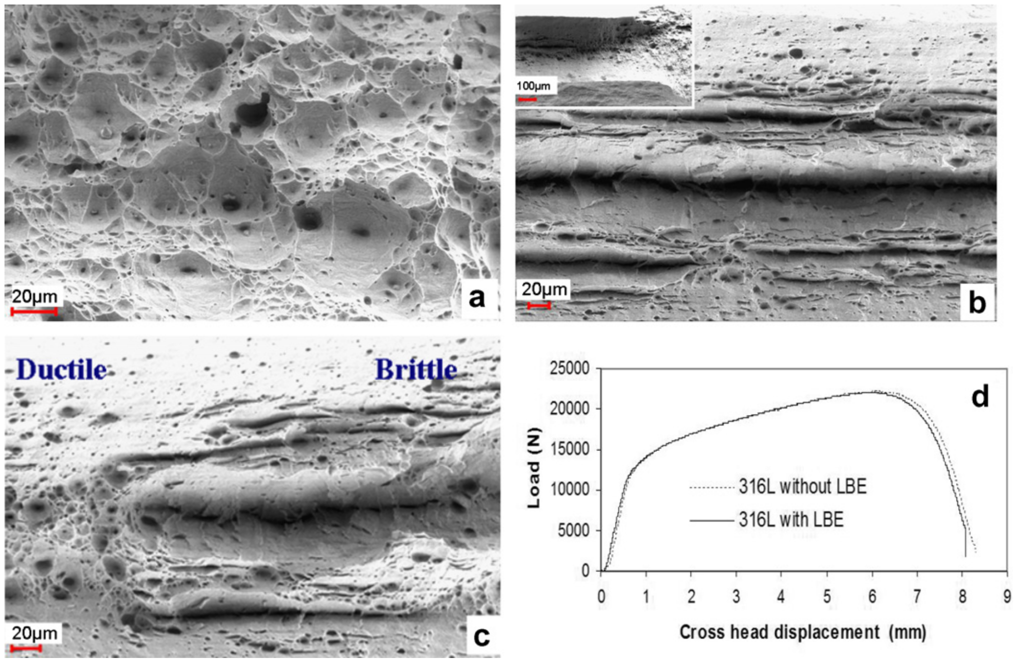

Fig. 3. 160 °C and 6.67×10^{-7} m s^{-1} (a) SEM micrographs of the fracture surfaces of the 316L CCT specimens, ductile in He–4%H_2 (reference test), (b) and (c) mixed brittle–ductile in LBE. The crack initiation zone is reproduced in inset of (b), (d) load according to cross-head displacement obtained on 316L with and without LBE.

mechanical behaviour of the material tested in the two different environments.

4. Discussion

The embrittlement effect of the LBE on T91 steel is very pronounced, which confirms the results of previous studies in other test conditions [10,18,19]. In the testing conditions of this paper, the fracture mechanism seems to be shear decohesion rather than quasi-cleavage in other studies [10,18]. This is most likely due to the plane stress condition of the chosen geometry. In such case, plastic deformation occurs at 45° of the loading axis.

Another important point resulting from this work is the ductility recovery at the highest investigated displacement rate, which suggests the existence of a brittle–ductile transition depending on the deformation rate and most likely also on the temperature. An opposite behaviour would be observed if cleavage was the prevailing mechanism of embrittlement, which is completely excluded in our case by the fractographic analysis. Following Rostoker et al. [13], the ductility recovery behaviour at the highest rate could be due to an ‘incubation time’ necessary for cracks to nucleate and propagate.

In the case of the 316L steel, a modification of the fracture surfaces, in contact with LBE, is reported for the first time. Such a modification competes with the normally ductile fracture. The mechanism of shear decohesion seems active but does not promote an embrittlement in a strict mechanical sense. A ductile–brittle transition is not excluded in the case of the 316L, since there is a very similar effect to that of T91. Therefore we can suppose that the applied conditions (T, deformation rate) do not allow the generalization of the effect; deformation rate can be too high to observe the full manifestation of the phenomenon.

For both steels, grain boundaries are not playing any role in crack branching (see Fig. 2(a) and Fig. 3(b)). Consequently, a mechanism independent of alloy microstructure can be used to describe the T91 and 316L steels embrittlement process by LBE. Most cases of LME can be attributed to the effects of chemisorption [4,5]. The diffusion and dissolution based models cannot account for the fastest crack growth rate because of the very low mutual solubilities of solid and liquid metals. It was recently shown that the solubility limit of iron in LBE is of 9×10^{-7} mol/cm^3 at 450 °C [20]. So only a phenomenon of adsorption can occur at the crack tip during fracture in contact with LBE. The adsorbed atoms at the crack tips can promote dislocation emission and even decohesion, depending on the fracture mode [21]. Therefore, it is tempting to invoke such a model to explain our findings. It would be interesting to study basic specimens coming from crack tips by the transmission electronic microscopy (TEM) to confirm these assumptions.

5. Conclusion

The effect of liquid LBE on the T91and 316L has been investigated by varying the deformation rate from 6.67×10^{-8} m s^{-1} to 6.67×10^{-3} m s^{-1}. The fracture proceeds by intense shear decohesion. The analysis performed with careful examination of the fracture surfaces, reveals that the degree of embrittlement depends on the strain rate. For T91, a transition from brittle to ductile fracture is found in the high strain rate range investigated. In the transitory stage, there is a competition between the growth of dimples and cracking induced by the liquid metal. For the 316L steel, the embrittlement effect is not pronounced. The fracture surfaces are characterized by a mixed brittle/ductile fracture profile. Concerning 316L, it will be interesting to explore other conditions (*T*, deformation rate); the applied deformation rate can be too high to have a generalized effect.

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References

[1] A. Alamo, J.L. Bertin, V.K. Shamardin, P. Wident, J. Nucl. Mater. 367–370 (2007) 54.
[2] A. Aiello, M. Azzati, G. Benamati, A. Gessi, B. Long, G. Scaddozzo, J. Nucl. Mater. 335 (2004) 169.
[3] M.H. Kamdar, The Metals Handbook 13 (1987) 171.
[4] M.H. Kamdar, Prog. Mater. Sci. 15 (1973) 289.
[5] N.S. Stoloff, T.L. Johnston, Acta Metall. 11 (251) (1963).
[6] J. Van den Bosch, D. Sapundjiev, A. Almazouzi, J. Nucl. Mater. 356 (2006) 237.
[7] V. Coen, P. Fenici, H. Kolbe, L. Orecchia, T. Sasaki, J. Nucl. Mater. 110 (1982) 108.
[8] D. Kalkhof, M. Grosse, J. Nucl. Mater. 318 (2003) 143.
[9] G. Nicaise, A. Legris, J.B. Vogt, J. Foct, J. Nucl. Mater. 296 (2001) 256.
[10] T. Auger, G. Lorang, Scripta Mater. 52 (2005) 1323.
[11] E. Pereiro-Lopez, W. Ludwig, D. Bellet, C. Lemaignan, Acta Mater. 54 (2006) 4307.
[12] B. Joseph, F. Barbier, G. Dagoury, M. Aucouturier, Scripta Mater. 39 (1998) 775.
[13] W. Rostoker, J.M. McCanghey, H. Markus, Embrittlement by Liquid Metals, Reinhold Publishing Corporation, Van Nostrand, New York, 1960.
[14] B. Joseph, M. Picat, F. Barbier, Eur. Phys. J. Appl. Phys. 5 (1999) 19.
[15] P.J.L. Fernandes, D.R.H. Jones, Eng. Fail. Anal. 4 (1997) 195.
[16] I. Ueda, M. Miyake, M. Kawamura, Trans. Jap. Weld. Soc. 4 (1) (1973).
[17] N.K. Batra, J.B. See, T.B. King, Weld. Res. J. 417s (1974) 417.
[18] A. Legris, G. Nicaise, J.B. Vogt, J. Foct, J. Nucl. Mater. 301 (2002) 70.
[19] Y. Dai, B. Long, F. Groeschel, J. Nucl. Mater. 356 (2006) 222.
[20] L. Martinelli, mécanisme de corrosion de l'acier T91 par l'eutectique Pb–Bi utilisé comme matériau de cible de spallation, PhD thesis, Université Paris VI, 2005.
[21] S.P. Lynch, Scripta Metall. 18 (1984) 509.