SEM STUDY OF FERRITIC-MARTENSITIC AND AUSTENITIC STEELS IRRADIATED IN CONTACT WITH LEAD BISMUTH EUTECTIC

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1. Introduction

Ferritic-martensitic steels are candidate materials for the containers of liquid targets of the spallation neutron source and accelerator driven systems facilities due to their high strength at elevated temperatures, low thermal stress and anticipated low liquid metal corrosion rates [1, 2]. Among the different types, the 7– 9 wt. % Cr martensitic grades are considered better than those martensitic steels with higher (10 – 13 wt.%) chromium contents because of their lower ductile–brittle transition temperature (DBTT) shifts after irradiation [3, 4].

The austenitic 316L is one of the candidates of the structural materials for accelerator driven systems and Generation IV reactor conceptual designs. Even having quite good corrosion resistance this material is sensitive to irradiation damage in the temperature range of 150–400 °C: even low levels of irradiation exposure, as small as 0.1 dpa, can cause severe loss of ductility during tensile loading. However, in this case, the corrosion of 316L in liquid alloy environment can be substantial, especially when a dissolution process occurs [5].

In order to understand the dissolution process of potential material's candidates for Accelerator Driven Systems (ADS), taking into account that the operational temperature of Lead Bismuth Eutectic (LBE) spallation target in the proposed ADS system is expected to be about 400 °C, the irradiation program (within EUROTRANS project) was launched to characterize materials irradiated in contact with LBE, the coolant and spallation target in ADS systems [6]. The Irradiation experiment of the Lead BIsmuth structural materials System (IBIS) has been performed in the High Flux Reactor (HFR) in Petten with the objective to investigate the synergistic effects of irradiation and LBE exposure on the mechanical properties of ferritic-martensitic T91 and austenitic 316L stainless steel [7]. The IBIS experiment was designed to be complementary to the irradiation experiment TWIN ASTIR in the BR2 with respect to irradiation temperature [8]. Corrosion and liquid metal embrittlement of 316L and T91 exposed to LBE has been reported to be controlled by the oxygen content and the degree of surface wetting with LBE, which depends on the exposure temperature as well [8-10]. The presented data are dealing with Scanning Electron Microscopy (SEM) study of LBE and irradiation embrittlement above mentioned materials.

2. Experimental details

The IBIS irradiation experiment was performed at two different irradiation temperatures: 500 and 300 ºC. Besides the irradiated containers, another identical reference container including the specimens was fabricated to test the influence of LBE on the materials without irradiation, the so called ‘0 dpa’ capsule. This capsule has experienced a similar temperature cycle as the capsule irradiated at 300 ºC. The capsules contain different types of T91 and 316L corrosion specimens. Corresponding chemical compositions are given in Table 1 [10].

The LBE used in the experiment was provided by Hetzel Metalle GmbH: 55.2 wt. % Bi and 44.8 wt. % Pb containing (2 mg/g Cr and less than 1 mg/g Ni). The specimens were subjected to fast fluences (E > 1 MeV) varying between 9.54 x 1024 m^{-2} and 16.7 x 1024 m^{-2}, with corresponding damage level ranges between 1.3 and 2.4 dpa for T91 specimens. The duration of irradiation was 250 full power days (FPD). The temperature was measured during irradiation at three vertical levels in each capsule (six levels in total) by means of 12 thermocouples. The original oxygen content in LBE during filling of the capsules was kept as low as 10^{-6} wt. % (which can be in reality even lower due to effect of time and temperature), which is needed to prevent corrosion.

The samples for SEM investigation were prepared from the irradiated specimens at the NRG Hot Cell Laboratories (HCL). The Au-Pd coating was used to facilitate conductivity of the surface to the holder. For the chemical analyses (EDS/WDS), this was taken into account in such a way that the amount of Au was always checked during measurement. Oxides were not considered in this study due to the low oxygen content present during the irradiation process [11].

Tab. 1. *Chemical composition of T91 and 316L steels used in IBIS experiment.*

Steels	Elements (wt. %)											
	Fe	C	Mn	P	Si	Ni	Cr	Mo	N	V	Cu	Nb	S
T91	balance	0.100	0.400	0.020	0.230	0.100	9.00	0.900	0.044	0.210	0.06	0.06	-
316L	balance	0.019	1.81	0.003	0.67	10	16.7	2.05	0.029	-	-	-	0.0035

3. Results and Discussion

In the Figure 1 the overview of the T91 and 316L SEM images is given. According to the obtained result (for more details see [12]), the chemical interaction between the irradiated steels and LBE seems to be stronger for 316L type of steel. This could be caused by higher content of Ni which makes this kind of steel more sensible to LBE influence [13]. It should be noticed that in our case the WDX analyses were showing more “fluctuation” of the amount of Ni at the boundaries “steel- LBE” then particular trend.

Many pores can be found in the interface between 316L steel and LBE even in the case of reference capsule. It was showed that these surface defects could further increase susceptibility to LBE and favour wetting [14].

The big pores can be found in the interface part of the 316L steel irradiated at 300 °C and this interface is not as sharp as for T91.

In the case of 500 °C irradiation interaction steel- LBE seems to be more advanced as expected. The T91 steel is showing complete wetting on same places, both, the 316L and T91 are showing penetration of LBE into the sample.

Generally two processes should be taken into account in the consideration of the reaction between steel and LBE. The first process is dissolution of the steel and subsequent ferritic layer formation. The second process is oxidation itself.

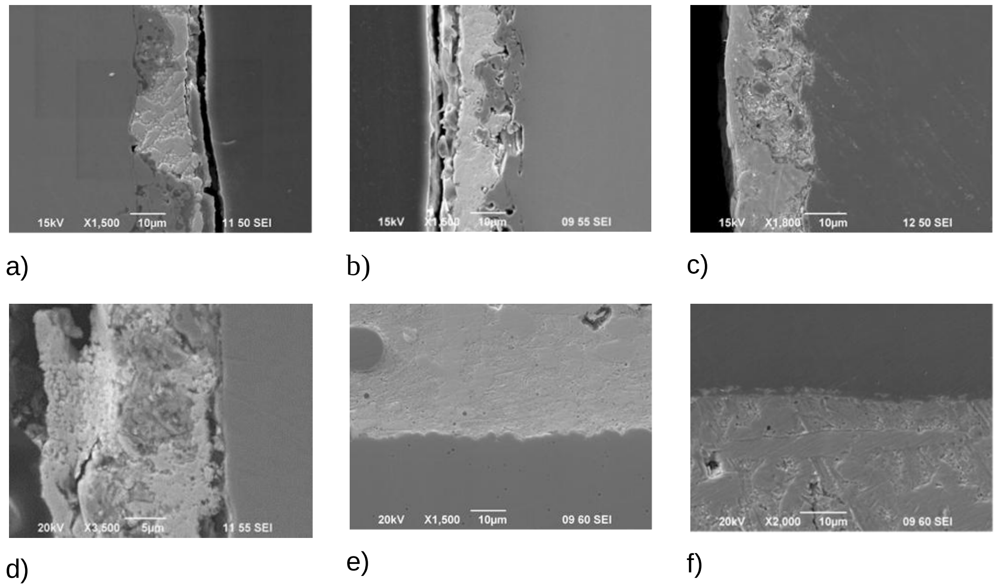

Fig.1: *SEM images of the reference specimens T91 (a) and 316L (b), specimen irradiated at 300 ^{o}C T91 (c) and 316L (d) and specimen irradiated at 500 ^{o}C T91 (e) and 316L (f).*

It was showed [15] that the 316L can be damaged by dissolution in low oxygen concentration containing LBE. At 450 °C Ni is preferentially dissolute (generally it is starting at 500 °C for most of the elements). The depletion of Ni, Mn and Cr is found in the ferritic layer on the sample's surface. Due to the dissolution the layer is porous and easy to be filled with liquid metal. There is not diffusion zone between metal and material and the composition is changed in the one step. It is also well known that oxygen concentration in liquid metal is a key parameter for corrosion of structural materials. Low oxygen content cause decreasing of dissolution resistance. According to the literature, down to 10^{-7} wt.% at 550 °C (for 3000 hours), the dissolution occurs. For higher oxygen content corrosion is taking place [13].

Summarizing above mentioned facts there are two possible reasons for stronger reaction of the steel at 500 °C i.e. temperature increase to more than 550 °C or lower oxygen content than expected. The amount of oxygen was aimed to be 10^{-6} wt%. The final concentration of oxygen in the capsule, however, can differ from aimed value, as it depends on the time when a thermodynamic stability of the system is reached. This thermodynamic stability is defined as an equilibrium between the dissolution of oxygen in the LBE and the reduction or oxidation of the oxide film on the stainless steel surfaces. Moreover, the solubility of oxygen and other chemical elements in the LBE is not constant during the irradiation experiment due to the fluctuations in temperature of the LBE caused by the cycles of the reactor. Therefore the final oxygen concentration is therefore difficult to predict and thus can be also lower (higher) than aimed value [16].

The temperatures were measured during irradiation at three vertical levels in each capsule (six levels in total) with 12 thermocouples. At every level two thermocouples were used, one in the central tube and the other in direct contact with the outside container tube. The average temperatures measured during irradiation varied between 290 and 319 °C for the 300 °C temperature section and between 478 and 489 °C for the 500 °C temperature section. Temperature control was achieved by the design of the gas gap between the containers and the specimen holder and by movement of the experiment in its core position [17].

In the IBIS experiment there were also tensile and KLST samples irradiated in LBE in order to follow (synergetic influence of LBE and neutron irradiation at different temperatures on the mechanical properties of the T91 and 316L steels and their welds. By Kolluri [17] it was showed that the damages induces by LBE interaction, observed by SEM and reported here are limited to several microns within the surface of the material and hence its influence on tensile properties is not visible. This together with the reading of the temperature and the fact that in the 500 °C container the significant blackening of the samples were observed can indicate that if the temperature increase really happened it was just locally on the surface of the specimens

4. Conclusions

The SEM observation showed that even in the case of reference samples the chemical interaction between the matrix and LBE took place. In both cases, reference samples and low temperature irradiation, the influence of LBE seems to be more significant for 316L steel in comparison with T91 steel.

In the case of high temperature irradiation (500 °C) up to 2.4 dpa the interaction between LBE and steel is more advanced. The T91 steel reveal porous surface of the matrix and completed wetting almost along whole surface of the sample. The 316L sample showed on same interaction places complete wetting and on some mixture of steel with LBE. Both kinds of steels showed small cracks filled by LBE at the interface whilst they occurrence is higher for T91 steel.

The EDS/WDS measurements did not reveal any trends of elements depletion close to the interface steel/LBE, only the variation in the concentration of Cr and Fe content were observed.

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