T91 STEEL IRRADIATED IN PB-BI ENVIRONMENT

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1. Introduction

Martensitic steels are candidate materials for the containers of liquid targets of the spallation neutron source and Accelerator Driven Systems (ADS) 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 of martensitic steels, the 7– 9 wt.% Cr grades are considered better than steels with higher (10–13 wt.%) chromium contents because of their lower ductile–brittle transition temperature (DBTT) shifts after irradiation [3,4].

In order to understand the dissolution process of potential candidate materials for ADS systems, 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. The program aims to characterize structural materials irradiated in contact with LBE, being the coolant and spallation target in ADS [5]. The Irradiation 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 9 wt.% Cr steel (T91) [6]. The corrosion and liquid metal embrittlement of 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 [7-9]. This work summarizes the results of obtained on pre-stressed T91 steels irradiated in LBE environment.

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. The 0 dpa capsule has experienced a similar temperature cycle as the capsule irradiated at 300 ºC. The capsules contain different types of T91 corrosion specimens: steels, welds, unstressed strips and bent strips. Corresponding chemical compositions is 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.

Tab. 1. *Chemical composition of T91 steel used in IBIS experiment (in wt.%).*

Elements	Fe	C	Mn	P	Si	Ni	Cr	Mo	N	V	Cu	Nb
m (wt.%)	balance	0.100	0.400	0.020	0.230	0.100	9.00	0.900	0.044	0.210	0.06	0.06

The samples for SEM (Scanning Electron Microscopy) investigation were prepared from the irradiated specimens at the NRG Hot Cell Laboratories (HCL). In the case of strips, they were first unscrewed from the holder and then cut in the middle. The cross-section was studied by SEM. Either silver paint or 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].

3. Results

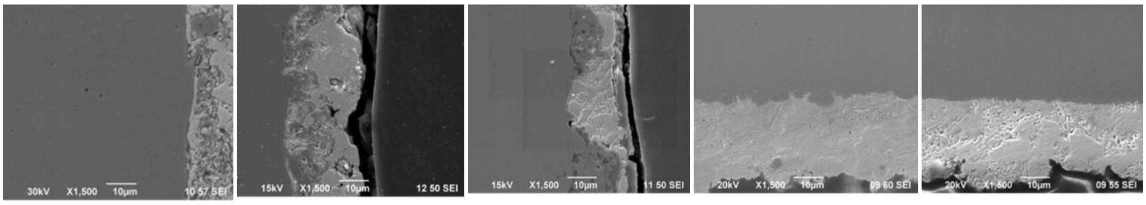
In the Figures 1 and 2 the overview of the T91 SEM images is given. The reference specimen was in contact with LBE at the temperature of 300 ºC for 250 days without irradiation. Some chemical interaction between the steel and LBE as well as pores in many instances are observed. The matrix did not reveal porosity. The WDS analyses of T91 specimen showed that slight depletion of steel from Cr close to the interface with LBE can be observed on some places (from original 9.29 wt % to 3.5 wt% in the close neighbourhood of the interface area). In the area of interaction the amount of Cr is very low, almost zero. The layer of LBE attached to the specimen shows a porous structure. The results clearly showed that the wetting of the steels by LBE is not complete even though the interaction is quite strong.

The so called irradiation at low temperature was performed at 300 ºC up to 1.33 dpa in contact with LBE for 250 days. The SEM images of the steel/LBE interface are presented in Figure 1 (b). Chemical interaction between the steels and LBE is clearly observed, although wetting with LBE is not complete. No pronounced traces of additional damage or pores in the inner surface of the samples are observed. SEM WDS spectra showed varying amount of Cr and Fe close to the boundary with LBE. In some places the content of Cr and Fe decrease in the vicinity of the boundary of the steel with LBE but never come to zero value.

The SEM results on similar T91/T91 welds submitted to low temperature irradiation are presented in Figure 1 (d). Rather extensive chemical interaction between the weld and LBE is observed in a number of locations. In multiple instances bubbles exposed to the surface are visible. WDS spectra were taken on different locations of the sample showed the changes in the amount of chromium and Fe at the vicinity of interface with LBE i.e. depletion of the Cr and Fe in the steel close to the boundary with lead-bismuth (from around 9.3 wt% to 7.5 wt%).

The so called irradiation at high temperature was performed at 500 ºC up to 2.44 dpa in contact with LBE for 250 days. The SEM result of T91 specimen are presented in Figure 1 (c). From the figure the interaction LBE with steel is clearly visible although the wetting is not complete. Porosity of the sample is observed and is less significant in comparison with the samples irradiated at 300 °C. From figure 1 (c) is seen that the LBE underwent restructuralization during exposure.

The WDS analyses showed only a slight decrease of Cr content (1-2 wt%) together with Fe (1-3 wt%) in the area in vicinity of LBE. The amount of Ni increases on some places at the boundaries of LBE and T91 (about 2 wt%). The SEM result of T91/T91 similar weld specimen is presented in Figure 1 (e). Complete wetting of the specimen with LBE is observed along the whole steel-eutectic interface. The intensive cracks are observed on the surfaces between LBE and steel. The morphology of the bulk of the LBE has changed compared to un-welded sample. This new morphology is characterized by 10-20 µm grains surrounded with areas with high porosity. The WDS analyses of M256 sample showed on multiple locations increased content of Ni in LBE in comparison with original composition (of about 3 wt%).



a) b) c) d) e)

Fig.1: *SEM image of (a) T91 reference, (b)T91 irradiated at 300 ºC and (c) T91 irradiated at 500 ºC, together with T91/T91welds (d) irradiated at 300 ºC and (e) irradiated at 500 ºC*

4. Discussion

Some significant surface roughness of steel can be found close to the interface between steel and LBE originated from preparation process. It was already proven that these surface defects could further increase susceptibility to LBE and favours wetting [12]. In the case of 500 °C irradiation, the steel-LBE interaction seems to be more notable as expected. The T91 steel is showing complete wetting in the some places and penetration of LBE into the sample via surface roughness, which could be even pre-existing one. Generally two processes should be taken into account when considering the reaction between steel and LBE. The first process is dissolution of the steel and subsequent ferritic layer formation. The second process is the oxidation itself. It is also well known that oxygen concentration in the liquid metal is a key parameter for corrosion of structural materials. The low oxygen content causes decrease of the dissolution resistance. According to the literature, down to 10^{-7} wt. % at 550 °C (for 3000 hours), a dissolution process occurs. For higher oxygen content corrosion takes place [11]. Summarizing the above mentioned facts, there are two possible reasons for stronger reaction of the steel with the LBE at 500 °C: namely the i.e. temperature increased above 550 °C or the oxygen content was lower then expected. The amount of oxygen in LBE was aimed to be 10^{-6} wt%. The final concentration of oxygen in the irradiation capsule, however, can differ from the target value, as it depends on the time when a thermodynamic stability of the system is reached. This thermodynamic stability is defined as 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 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 target value [13]. According to the data from thermocouples mentioned in experimental part, the more probable is that the dissolution at 500 ºC was caused by low oxygen content in LBE than by overheating during irradiation experiment.

5. Conclusions

The SEM observation showed that even in the case of reference samples the chemical interaction between the matrix and LBE took place. The T91/T91 weld irradiated at 300 °C to 1.3 dpa reveals many pores in the matrix surface. 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 almost completed wetting. The T91/T91 weld has intensive cracks on the surfaces between LBE and steel. In some places complete wetting is achieved. The EDS/WDS measurements did not reveal any trends of elements depletion close to the interface steel/LBE. Just variation in the concentration usually in Cr and Fe content were observed.

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