SEM CHARACTERIZATION OF PRE-STRESSED 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: 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. In the case of pre-stressed strips, the irradiation induced stress relaxation was measured (the change of the curvature before and after release of the screws) before cutting. For the SEM investigation, 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.

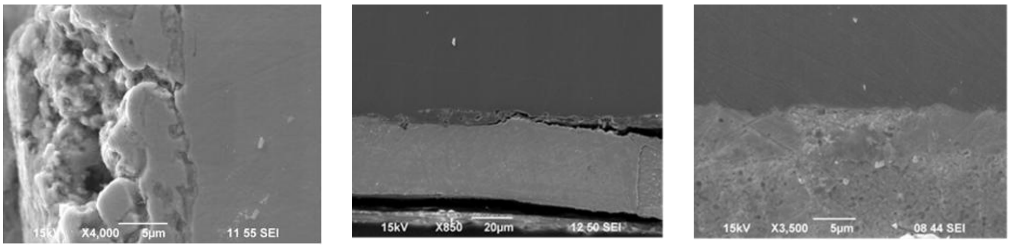
3. Results

The T91 pre-stressed sample (Figure 1 a) was in contact with the LBE at 300 ºC for 250 days without any irradiation. The sample exhibits some highly damaged areas with a high amount of surface roughness due to sample preparation. Nodirect contact LBE layers is observed in those areas (see Figure 1 a). The bulk of the sample is almost free of pores.

The T91 pre-stressed sample in Figure 1 b has been irradiated in LBE at 300 ºC for 250 days up to 1.3 dpa. The LBE layer is disconnected from the sample in the middle where the mechanical stress is supposed to be the highest (see Figure 1 b). In this area, also the LBE exhibited a specific coarse granular structure. Small surface roughnesses filled with LBE are observed in the area of contact between LBE and T91 steel. The outer surface of the sample in the area of higher stress seems to be highly damaged.

The SEm results of T91 pre-stressed sample irradiated in LBE at 500 °C for 250 days up to 2.4 dpa. are presented in Figures 1c. The layer of LBE is present along the whole exposed surface. Full wetting is observed along the steel/LBE interface. Small surface roughnesses filled with LBE are observed in the area of contact between LBE and steel.

The results of WDS analysis demonstrate slight decrease of the Cr content (2-4 wt.%) in the area in vicinity of the steel/LBE interface, whilst 2 wt.% change is typical for the higher irradiation temperature [11]. In the case of non-irradiated sample the variation is negligible. The radiation induced relaxation was calculated using the pre-stressed samples. The lower curvature and residual stress value (157 MPa) correspond to the sample irradiated at 300 °C to 1.3 dpa (Table 2). This could be related to the pronounced corrosion layer at the sample irradiated at 500 °C (residual stress about 189 MPa).



a) b) c)

Fig.1:*SEM image of reference pre-stressed T91 specimens; (a) reference, (b) irradiated at 300 ºC and (c) irradiated at 500 ºC.*

Tab. 2.*Results of radius calculation for experimental data from T91 pre-stressed corrosion strips irradiated at different temperatures to different doses together with residual stress data..*

T91 Sample	Properties			
	Radius (ϱ_0) of the strip bend on the holder [mm]	Radius (ϱ_1) of the strip [mm]	Applied stress (σ_0) [MPa]	Residual stress (σ) [MPa]
Reference sample	1	2	3	4
1.3 dpa, 300 °C	5	6	7	8
2.4 dpa, 500 °C	9	10	11	12

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 sample, chemical interaction (compositional changes) between the matrix and LBE occurred. In the case of high temperature irradiation (500 °C) up to 2.4 dpa, the interaction between the LBE and steel is more significant. The T91 steel reveals porous surface of the matrix and complete wetting almost along the whole surface of the sample. The pre-stressed T91 sample irradiated at 300 °C to 1.3 dpa showed the lower curvature as well as lower residual stress then the sample irradiated at higher temperature.

The EDS/WDS measurements do not reveal any trends of elements depletion close to the steel/LBE interface. Mostly, only variation in Cr content is observed.

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